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CATALYTIC TWO-STAGE LIQUEFACTION OF NOVA SCOTIA COALS, PART II

FINAL REPORT

by

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SUMMARY

Part II of a research program on catalytic two-stage liquefaction of Nova Scotia bituminous coals has been completed. Test work was focused on catalyst performance and coal space velocity in the first-stage reaction zone. Experiments were carried out in a once-through processing mode using a continuous-flow bench unit.

During the first phase of work on catalytic two-stage processing a nickel-molybdenum bimodal catalyst, Amocat 1C, was tested in both stages. On completion of this work a bimodal catalyst with higher nickel-molybdenum loading and larger mean mesopore and macropore diameters, Harshaw/Filtrol 2, was tested in the first-stage reactor. A total of ten experiments were carried out with the Harshaw/Filtrol catalyst using three Sydney area coals and Illinois No. 6, a benchmark American coal. High coal conversions, 90-95%, were achieved with all four coal feedstocks at a first-stage temperature of 400°C. Lingan Harbour, Donkin Harbour and Prince Hub coals produced 38, 45 and 49% oil respectively compared to 55% from Illinois No. 6. Amocat 1C was found to be superior to Harshaw/Filtrol 2 in terms of activity and stability. The superior performance of Amocat 1C is primarily attributed to its high surface area in combination with suitable type and adequate amount of catalytic metals.

After completion of experimental work with the Harshaw/Filtrol catalyst a larger first-stage reactor was installed in the bench unit to provide one-half the slurry space velocity employed in earlier tests. Eleven experiments were conducted with Amocat 1C to assess coal reactivity and the effects of reaction temperature and catalyst age. Coal conversion ranged from 93 to 94% with the three Sydney area coals. In experiments at similar operating conditions and catalyst age, oil yields from Lingan, Donkin and Prince coals were 47, 51 and 55% respectively. Improved process performance was observed at low coal space velocity with fresh, highly-active catalyst. However, test results suggest that longer residence time increases the rate of catalyst fouling over the range of coal space velocity investigated.

Additional work on advanced two-stage liquefaction is planned. The overall objective is to define the optimum two-stage process configuration for high rank coals from the Sydney coalfield. Laboratory efforts will be concentrated on asphaltene conversion studies and catalyst regeneration and activity testing. Bench unit operations will continue to assess the performance of promising new and/or reactivated catalysts.

INTRODUCTION

In Canada, our way of life is highly dependent on petroleum liquids. Since our existing crude supplies are not expected to meet our future liquid fuel demands, it seems that we will have to rely on synthetic liquid fuels to some degree in the next few decades. Thus, should it be economically attractive or essential, abundant coal reserves are available in Canada for conversion to synthetic liquid fuels. Because long lead times are involved in process evaluation, detailed design and plant construction it is essential to understand the basic concepts of the promising liquefaction processes and verify their potential yield structure as they apply to Canadian coals.

The in-situ coal resources of Nova Scotia occur in five main fields: Sydney, Mabou, Port Hood, Pictou and Springhill. The largest resources, over 2 billion metric tons, occur in the submarine area of the Sydney coalfield. The coal-bearing section of the Sydney basin contains eleven major seams ranging from 1 - 4.5 m thick. Historically, the two most productive seams have been the Harbour and Phalen seams.

At present, the principal coal producer in the Province is the Cape Breton Development Corporation with two operating mines, Lingan and Prince, and a third under development at Donkin. All three mines are located in the Sydney area. The Harbour seam is currently being worked at Lingan whereas Hub seam coal is being produced from the Prince mine. Both coals are classified as high volatile A bituminous.

Process studies relating to the direct liquefaction of Nova Scotia high volatile bituminous coals were initiated at the Nova Scotia Research Foundation Corporation (NSRFC) in 1978. Initially, efforts were concentrated on a low-severity extractive coking process (1, 2, 3). From 1980 to 1984, experimental work was conducted on medium to high pressure donor extraction (4,5) and single-stage catalytic hydrogenation (6). Yield data has been developed over a wide range of operating conditions using bench-scale batch and continuous-flow hydroprocessing units. The product slate of interest is liquid transportation fuels.

Test results have shown that single-stage catalytic hydrogenation produces higher oil yields than donor solvent extraction. Furthermore, it has been shown that considerable control can be exercised over product distribution by varying process severity. Once-through experiments with Lingan Harbour seam coal and commercial nickel-molybdenum hydrotreating catalyst over medium to high severity operating conditions produced high coal conversions, 90 - 92%, while oil yields ranged from 30 - 47%.

Direct catalytic coal liquefaction processes, such as H-Coal, are single-stage processes. The catalyst serves several functions and the severe operating conditions required for one function might not be needed for other functions. Recent process developments in the United States are centered on liquefying coal in two separate reaction zones. Two-stage processing offers the versatility of controlling coal dissolution and primary product upgrading under different operating conditions.

During the first half of 1985 our bench-scale coal liquefaction unit was modified and expanded for integrated two-stage operations. The closely coupled two reactor system offers the potential of higher distillate yields and improved hydrogen utilization. From mid-1985 to late 1986 the unit was operated with a first-stage thermal dissolver and a second-stage catalytic reactor. High coal conversions, 92 - 94%, were achieved in once-through experiments with Lingan Harbour, Donkin Harbour and Prince Hub seam coals using alumina base hydroprocessing catalysts in the second-stage reactor (7,8). In a six-day recycle liquefaction test with Prince coal and a nickel-molybdenum bimodal catalyst the net C_4^+ liquid yield was 67% during the last twelve-hour operating period. The yields of naphtha and light gas oil were 44 and 38 wt. % respectively (9).

The first-stage thermal dissolver was converted to a catalytic reactor in 1987 to undertake catalytic two-stage test work (10). Experiments conducted using a nickel-molybdenum bimodal catalyst in both stages produced higher oil yield, lower gas make and higher asphaltene/preasphaltene yield than the thermal/catalytic processing mode. Recently, bench-scale studies have been concentrated on catalyst performance and coal space velocity in the first-stage reaction zone. Results of this work are presented herein.

TASK 1 - EQUIPMENT MAINTENANCE

Mechanical work asscciated with this task began in June of 1987. Activities included inspection and general maintenance of the hydrogen compression facility, the fixed-bed catalytic hydrotreater and the two-stage coal liquefaction unit.

In the hydrogen compression and storage area scheduled maintenance was carried out on the single-stage diaphragm compressor. The design capacity of this 7.5 horsepower unit is 114 L/min from 3.5 MPa to 34.5 MPa. Routine maintenance involved cleaning filters, replacing the hydraulic fluid and checking the tension on drive belts. The triple diaphragm assembly and o-ring seals were also replaced during the maintenance period.

The fixed-bed hydrotreater used to produce reaction solvent for coal liquefaction experiments has been described in an earlier report (3). On turnaround, the high pressure liquid metering pump and the vacuum pump used regularly with this system were given a general overhaul. Liquid transfer lines between the feed vessel and the reactor inlet were removed and cleaned. Following this, new heating tapes and insulation were installed on these lines. In addition, high pressure block valves on both feed and product vessels were replaced due to wear associated with normal usage.

A complete description of the two-stage coal liquefaction unit is presented elsewhere (10). Most of the work associated with Task 1 was related to the mechanical maintenance of this unit. Inspections of both the first and second-stage stirred tank reactors revealed worn internal components. Worn parts, in particular the lower drive shaft and the bearing in the base weldment, were replaced in both reactors at NSRFC's machine shop. Several other parts on the rig were also replaced including: all rupture discs, worn high pressure block valves and frayed high temperature insulation. Product let-down valves were inspected and worn parts were replaced. Wet test meters were thoroughly cleaned and then performance tested.

TASK 2 - PROCUREMENT OF NEW EQUIPMENT

The purpose of Task 2 was to procure a new stirred tank reactor and filtration equipment to enhance our coal liquefaction capabilities.

The first item, a 3.8-liter stirred tank reactor complete with Robinson/Mahoney style internals was purchased from AE Autoclave of Canada Limited. The reactor internals include a catalyst basket and an impeller/baffle system. The double ended bolted closure reactor is designed with double delta ring gaskets. This reactor was used to replace the 1.5-liter first-stage reactor in the two-stage coal liquefaction unit. Test work was proposed with the larger reactor to study the effect of first-stage slurry space velocity on overall process performance.

Laboratory-scale batch pressure filtration equipment for solids/liquid separation of liquefaction bottoms product was purchased from Sepor Inc. of California. The recovery of liquid from separator bottoms product is an integral part of recycle coal liquefaction operations.

TASK 3 - PREPARATION OF RAW MATERIALS

3.1 Coal Feedstocks

Four coal feedstocks were tested in this study. Experimental work concentrated on three Nova Scotia high volatile A bituminous coals, Donkin Harbour, Lingan Harbour and Prince Hub. Limited test work was conducted on a high volatile American coal, Illinois No. 6. The Sydney area coals were provided courtesy of the Cape Breton Development Corporation. Illinois No. 6 coal from the Burning Star Mine was procured through Cepheus Industries Incorporated, Marion, Illinois. Preparation of the coals involved riffling, grinding and screening. After screening, the individual coals were thoroughly mixed and stored in plastic-lined drums with a nitrogen blanket.

Proximate and ultimate analysis of the coals are given in Table 1. Proximate analysis, free-swelling index and forms of sulphur were determined according to standard methods. Total sulphur was measured using a Leco sulphur analyzer. Carbon, hydrogen and nitrogen were determined using a Perkin-Elmer elemental analyzer.

Petrographic analysis of the four coals is given in Table 2. These analyses were carried out courtesy of Atlantic Geoscience Center, G.S.C., Dartmouth, Nova Scotia.

TABLE 1

ANALYSIS OF COALS TESTED

COAL DESCRIPTION:	LINGAN HR-9	DONKIN HR-9	PRINCE HB-9	ILLINOIS NO. 6
Moisture, Wt. %	1.08	2.51	3.19	2.91
PROXIMATE (Dry Basis) Wt. %				
Ash	4.18	5.65	7.16	9.81
Volatile Matter	35.23	37.03	37.36	40.97
ULTIMATE (Dry Basis) Wt. %				
Carbon	80.13	77.34	73.88	69.64
Hydrogen	5.45	5.28	4.95	4.94
Nitrogen	1.27	1.48	1.31	1.46
Sulphur	2.00	2.82	3.46	2.89
Ash	4.18	5.65	7.16	9.81
Oxygen (by difference)	6.97	7.43	9.24	11.26
SULPHUR FORMS (Dry Basis) Wt. %				
Sulphate	0.04	0.49	0.14	0.04
Pyritic	1.49	1.04	2.38	1.17
Organic (by difference)	0.47	1.29	0.94	1.68
Total	2.00	2.82	3.46	2.89
FREE SWELLING INDEX	7 1/2	7	6 1/2	2 1/2
HEATING VALUE (kj/kg)	34,162	31,880	30,393	28,652

TABLE 2

PETROGRAPHIC CHARACTERISTICS OF COALS TESTED

COAL DESCRIPTION:	LINGAN HR-9	DONKIN HR-9	PRINCE HB-9	ILLINOIS NO. 6
MACERAL COMPOSITION (% by volume)				
Vitrinite	73.2	78.4	78.4	75.6
Exinite	4.2	6.0	4.6	4.4
Inertinite	19.6	13.8	11.8	12.4
Macrinite	(2.8)	(1.8)	(1.4)	(1.2)
Micrinite	(4.2)	(3.4)	(1.6)	(2.2)
Semifusinite	(5.2)	(3.0)	(5.0)	(3.4)
Fusinite	(7.4)	(5.6)	(3.8)	(5.6)
Mineral Matter	3.0	1.8	5.2	7.6
Pyrite	(0.4)	(1.2)	(2.6)	(1.8)
Quartz	(0.2)	(0.2)	(0.2)	(1.4)
Shale	(2.4)	(0.4)	(2.4)	(4.4)
% R_0 Max.	0.98	0.91	0.74	0.60
V-Type Distribution, %				
V5	-	-	-	44
V6	-	-	22	56
V7	-	4	62	-
V8	10	30	16	-
V9	46	60	-	-
V10	42	6	-	-
V11	2	-	-	-

3.2 Anthracene Oil

This material is a coal-tar distillate which contains ca. 5.3 wt. % hydrogen. It was supplied by Domtar Chemicals Limited (Hamilton, Ontario). Hydrogenated solvent used in the two-stage coal liquefaction experiments was made from two batches of this highly aromatic oil.

The characteristics of the raw oil are presented in Table 3. Carbon, hydrogen and oxygen were determined using a Perkin-Elmer elemental analyzer. A standard kjeldahl method was used for the determination of nitrogen. Total sulphur content was measured by x-ray fluorescence.

In this study, the hydrogen aromatic/aliphatic distribution was established by proton nuclear magnetic resonance (H-NMR). A varian EM360L continuous wave spectrometer operating in the Fourier transform mode at 60 MHz, and a varian T60 NMR spectrometer operating in the Fourier transform mode at 60 MHz were used to obtain H-NMR spectra. Proton assignments were as follows: aromatic hydrogens (H_{ar}) were found in the region of 9 - 6 ppm. Benzylic hydrogens including methyls, occur at 3.5 to 2 ppm (H_α). Hydrogen, except methyls, on carbons one or more positions removed from the aromatic rings occur at 2 - 1.1 ppm (H_β). Aliphatic methyls are found at 1.1 - 0.4 ppm (H_γ). As almost no long aliphatic side chains are believed to be present, the H_β protons are predominantly in rings. These assignments are consistent with similar

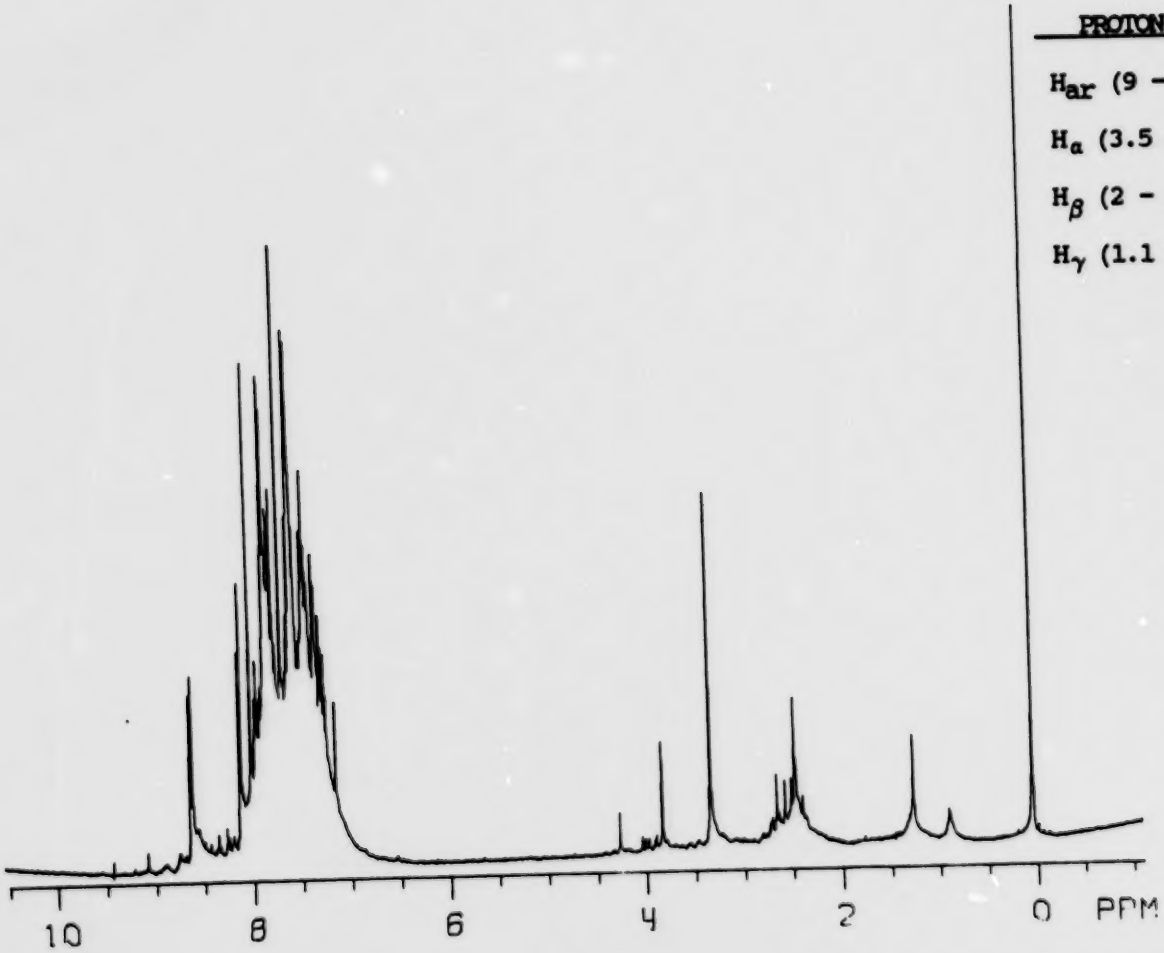
analyses reported in the literature (11, 12). Figure 1 is a typical H-NMR spectrum of raw anthracene oil.

TABLE 3

ANALYSIS OF RAW ANTHRACENE OIL

SAMPLE DESCRIPTION:	BATCH No. 8-19	BATCH No. 8-20
Specific Gravity, 60/60°F	1.1590	1.1607
Carbon, Wt. %	90.77	90.41
Hydrogen, Wt. %	5.27	5.25
Nitrogen, Wt. %	1.04	1.03
Sulphur, Wt. %	0.55	0.54
Oxygen, Wt. %	1.76	1.23
Proton Distribution, %		
H_{ar} (9 - 6 ppm)	88.09	88.29
H_{α} (3.5 - 2 ppm)	8.46	8.88
H_{β} (2 - 1.1 ppm)	2.17	1.73
H_{γ} (1.1 - 0.4 ppm)	1.28	1.10
Distillation Data, °C (ASTM D-1160)		
IBP	201	195
10%	323	320
20%	333	330
30%	344	344
40%	356	352
50%	366	361
60%	386	372
70%	402	384
80%	426	409
90%	456	442
95%	480	463
EP	523	502

Figure 1 - H-NMR SPECTRA OF RAW ANTHRACENE OIL WITH 5.25 WT. % HYDROGEN



PROTON DISTRIBUTION, %	
H_{ar} (9 - 6 ppm)	88.29
H_{α} (3.5 - 2 ppm)	8.88
H_{β} (2 - 1.1 ppm)	1.72
H_{γ} (1.1 - 0.4 ppm)	1.10

3.3 Catalysts

Three hydroprocessing catalysts were used in this study: Harshaw/Filtrol 2, Amocat 1C and Nalcomo 479. The Harshaw/Filtrol and Amocat catalysts are both alumina-supported $NiO-MoO_3$ formulations with bimodal pore size distribution. They were used in the two-stage coal liquefaction experiments. Nalcomo 479 is an alumina-supported $CoO-MoO_3$ catalyst with a unimodal pore size distribution. It was used to prepare reaction solvent. All catalysts were used in extrudate form. The chemical and physical properties of the three catalysts are given in Table 4.

TABLE 4

CHEMICAL AND PHYSICAL PROPERTIES OF HYDROTREATING CATALYSTS

	AMOCAT 1C	HARSHAW/ FILTROL 2	NALCOMO 479
MoO_3, Wt. %	16.0	18.9	19.0
NiO, Wt. %	3.0	3.9	-
CoO, Wt. %	-	-	4.4
Size, cm	0.16	0.16	0.16
Surface Area, M^2/g	190	129	180
Pore Volume, cc/g	0.85	0.66	0.47
Density, g/cc	0.61	0.60	0.82 - 0.84
Crush Strength, kg	4.4	-	6.4 - 7.3
Modal Pore Diameter, A^o	100/1500	166/3200	100

TASK 4 - HYDROGENATION OF ANTHRACENE OIL

Vehicle oil used in the Catalytic Two-Stage Liquefaction experiments was prepared from raw anthracene oil using a fixed-bed catalytic hydrotreater. The bench-scale unit has been described in an earlier report (3).

A commercial cobalt-molybdenum hydrotreating catalyst, Nalcomo 479, was used to prepare solvent containing 9% hydrogen. A summary of the operating conditions used to produce the hydrogenated oil is given in Table 5. The characteristics of two 9% H oil blends are shown in Table 6. An H-NMR spectrum of 9% H solvent is presented in Figure 2. Hydrogenated anthracene oil, 7.5% H, spiked with dimethyl disulphide was used to sulphide Harshaw/Filtrol 2 and Amocat 1C catalysts prior to catalytic two-stage testing in Task 5. The spiked oil was on hand from previous sulphiding runs.

Approximately 250 liters of 9% H reaction solvent was produced during this phase of work. The solvent was prepared at the rate of 1.8 liters per 7-hour shift. The catalytic hydrotreater was often operated two shifts per day. Daily production was collected to obtain 20-liter batches. Analysis of selected batches are presented in Table 7. Two composite blends of the 20-liter batches were made during this contract period. The analysis given in Table 6 is representative of the two oil blends.

TABLE 5

REACTION CONDITIONS FOR ANTHRACENE OIL HYDROGENATION

Reactor Temperature, °C	366 - 369
Reactor Pressure, MPa	15.51
LHSV, hr^{-1}	0.20
Hydrogen Flow, L/hr	175 - 295
Product Hydrogen Content, Wt. %	9.0

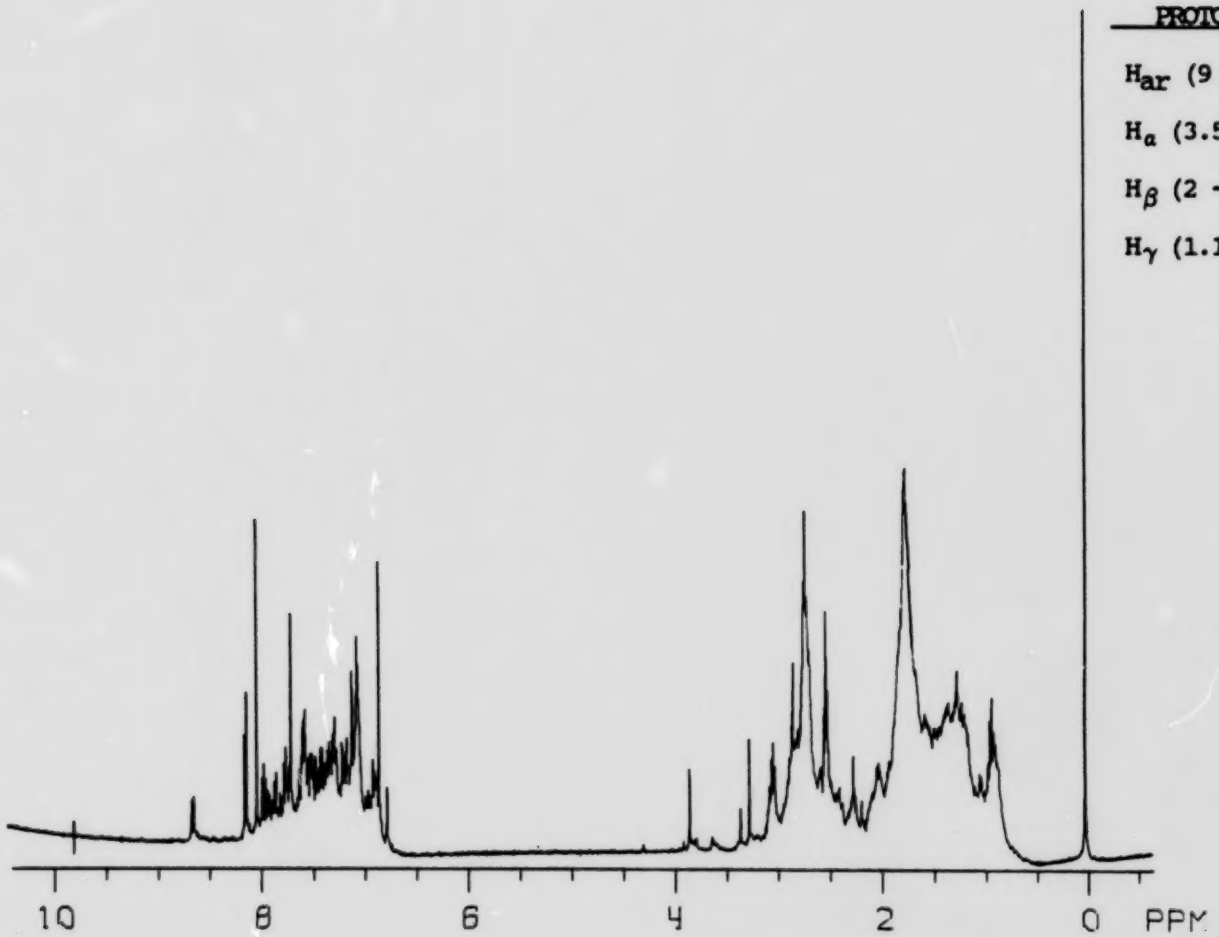
Catalyst used: Nalcomo 479

TABLE 6

CHARACTERISTICS OF HYDROGENATED ANTHRACENE OIL

SAMPLE DESCRIPTION:	HAO 91	HAO 92
Specific Gravity, 60/60°F	1.0341	1.0381
Carbon, Wt. %	89.89	89.95
Hydrogen, Wt. %	9.09	9.03
Nitrogen, Wt. %	0.16	0.16
Sulphur, Wt. %	0.008	0.009
Oxygen, Wt. %	0.85	0.87
Proton Distribution, %		
H_{ar} (9 - 6 ppm)	25.47	25.61
H_{α} (3.5 - 2 ppm)	26.26	26.79
H_{β} (2 - 1.1 ppm)	37.87	37.15
H_{γ} (1.1 - 0.4 ppm)	10.40	10.45
Distillation Data, °C (ASTM D-1160)		
IBP	212	195
10%	256	259
20%	285	280
30%	296	290
40%	304	298
50%	313	307
60%	322	315
70%	335	328
80%	354	346
90%	376	366
95%	388	385
EP	459	455

Figure 2 - H-NMR SPECTRA OF HYDROGENATED ANTHRACENE OIL WITH 9.03 WT. % HYDROGEN



PROTON DISTRIBUTION, %	
H_{ar} (9 - 6 ppm)	25.61
H_{α} (3.5 - 2 ppm)	26.79
H_{β} (2 - 1.1 ppm)	37.15
H_{γ} (1.1 - 0.4 ppm)	10.45

TABLE 7

ANALYSIS OF 20-LITER BATCHES OF 9% H SOLVENT

BATCH NO.	1	2	3	4	5	6
Specific Gravity, 60/60°F	1.0345	1.0355	1.0365	1.0374	1.0392	1.0364
Carbon, Wt. %	90.08	90.24	90.18	90.09	90.28	89.31
Hydrogen, Wt. %	8.97	9.01	8.98	9.04	9.02	9.03
Nitrogen, Wt. %	0.17	0.18	0.18	0.18	0.19	0.17
Sulphur, Wt. %	0.007	0.008	0.008	0.008	0.010	0.008
Proton Distribution, %						
H_{ar} (9 - 6 ppm)	23.86	24.54	24.61	25.61	25.38	24.36
H_{α} (3.5 - 2 ppm)	26.23	26.02	26.21	26.21	26.22	25.77
H_{β} (2 - 1.1 ppm)	40.13	35.93	38.44	37.45	36.82	38.52
H_{γ} (1.1 - 0.4 ppm)	9.78	13.52	10.74	10.73	11.58	11.35
Distillation Data, °C (ASTM D-86)						
IBP	165	170	169	162	175	140
5%	258	262	262	260	260	259
10%	275	280	279	278	281	279
20%	295	297	299	297	299	298
30%	310	310	310	310	312	310
40%	321	319	320	319	322	318
50%	330	328	330	330	332	330
60%	338	339	341	341	342	341
70%	350	350	354	354	355	353
80%	366	366	370	372	372	369
90%	389	388	395	398	396	391
95%	409	406	416	426	418	410
EP	517	498	502	502	516	507

TABLE 7 (Continued)

ANALYSIS OF 20-LITER BATCHES OF 9% H SOLVENT

BATCH NO.	7	8	9	10	11	12
Specific Gravity, 60/60°F	1.0378	1.0378	1.0374	1.0381	1.0384	1.0364
Carbon, Wt. %	89.89	89.96	89.92	90.08	89.97	90.13
Hydrogen, Wt. %	9.01	9.04	8.98	9.05	8.98	8.98
Nitrogen, Wt. %	0.17	0.18	0.18	0.16	0.17	0.17
Sulphur, Wt. %	0.008	0.008	0.010	0.010	0.010	0.011
Proton Distribution, %						
H_{ar} (9 - 6 ppm)	24.07	25.67	24.61	25.10	24.55	25.26
H_{α} (3.5 - 2 ppm)	26.00	27.80	26.18	27.33	26.35	27.25
H_{β} (2 - 1.1 ppm)	38.79	37.83	38.20	36.60	38.21	37.96
H_{γ} (1.1 - 0.4 ppm)	11.14	8.70	11.01	9.53	10.88	9.53
Distillation Data, °C (ASTM D-86)						
IBP	142	138	142	139	147	144
5%	256	255	254	255	253	258
10%	278	278	276	280	275	273
20%	297	296	295	297	296	294
30%	308	309	308	310	309	307
40%	319	319	317	320	319	316
50%	330	330	329	331	330	326
60%	341	340	340	341	341	338
70%	354	353	352	354	354	351
80%	369	369	370	370	370	367
90%	391	391	393	393	394	394
95%	410	411	416	415	415	418
EP	507	490	496	504	508	499

TASK 5 - CATALYTIC TWO-STAGE LIQUEFACTION

In two-stage liquefaction a catalytic first-stage reactor offers potential for lower severity coal dissolution than a thermal dissolver. The presence of an effective hydrogenation catalyst in the first-stage enhances the donor capacity of the recycle solvent and contributes to the stabilization of the initial products of coal dissolution.

Bench-scale studies on catalytic two-stage liquefaction of Nova Scotia coals were initiated at NSRFC during the first quarter of 1987. A limited number of experiments were carried out with Amocat 1C nickel-molybdenum bimodal catalyst in both reactors. The objective of Task 5 was to conduct further test work on this process scheme. The first priority was to evaluate the performance of a first-stage catalyst with higher metals loading and larger modal pore diameters than Amocat 1C. Following this, process performance was investigated as a function of slurry space velocity in the first-stage reaction zone.

A schematic diagram of the two-stage bench unit used is shown in Appendix E. The unit has been described previously (10).

A typical coal liquefaction run was of 10 hours duration. However, an additional 12 hours was required for preliminary and post run operations. In the majority of runs the controlled variables lined-out

about 4-5 hours after start-up. During the steady state operating period, vent gas streams were sampled and other product streams were collected for analytical and material balance purposes. A detailed operating procedure is given in Appendix E.

In the experiments described hereafter, the following definitions for the product fractions isolated by solvent extraction were used:

oil - pentane solubles

asphaltenes - toluene solubles, pentane insolubles

preasphaltenes - THF solubles, toluene insolubles.

A direct extraction technique was used to determine these fractions. Individual samples of the product were separately extracted with pentane, toluene and tetrahydrofuran (THF). Conversions were calculated on the basis of maf coal fed to the reaction zone during the steady state sampling period and unconverted coal in the dry insoluble residues. Conversion calculations are described in Appendix A.

Vent gas streams were analyzed using a Perkin-Elmer Sigma 3B gas chromatograph interfaced with a Perkin-Elmer Sigma 10 chromatography data station. Water collected in the high pressure knock-out vessel was analyzed for ammonia and sulphide using standard wet chemical methods.

5.1 Operations Using Harshaw/Filtrol Bimodal Catalyst

On completion of planned maintenance, the two-stage bench unit was reassembled with 0.1 kg of Harshaw/Filtrol 2 catalyst in the 1.5-liter first-stage reactor and 0.3 kg of Amocat 1C catalyst in the 3.8-liter second-stage reactor. The rig was then pressure tested at pressures up to 17.24 MPa at room temperature. Following this a trial run was made with oil alone to verify the operational status of mechanical and electrical systems.

Both Harshaw/Filtrol 2 and Amocat 1C are nickel-molybdenum bimodal catalysts. However, Harshaw/Filtrol 2 has higher metals loading and larger mean mesopore and macropore diameters than Amocat 1C as shown in Table 4. Prior to initiating coal liquefaction experiments, a 30-hour run was carried out to sulphide the fresh catalysts using hydrogenated anthracene oil, 7.5% H, spiked with dimethyl disulphide. The sulphiding procedure was conducted at 17.24 MPa and a start-of-run temperature of 175°C. Temperature was increased gradually over a 24-hour period to a maximum of 370°C.

A total of ten catalytic two-stage liquefaction experiments were completed under Task 5.1. Test work was concentrated on Harbour and Hub seam coals from the Sydney coalfield. One run was carried out with Illinois No. 6 coal for comparative purposes. The experiments were conducted in a once-through processing mode using hydrogenated anthracene

oil containing 9% H as reaction solvent. Coal concentration in the feed slurry was fixed at 28 wt. %. Hydrogen treatrate was ca. 20 wt. % based on dry coal.

The first liquefaction experiment, Run No. 9000, was of 24 hours duration using Lingan Coal to condition the catalyst. The run was carried out at a slurry feedrate of 1.5 kg/hr and a total pressure of 17.24 MPa. First and second-stage reactor temperatures were 415 and 425°C respectively. Details pertaining to operating conditions and performance are presented in Appendix B. Material balance data is given in Appendix C.

After completion of the catalyst conditioning operation, a series of experiments were conducted to investigate coal reactivity and the effects of reaction temperature, pressure and catalyst age on raw product yields. A summary of the results from these experiments is shown in Table 8.

TABLE 8

RESULTS FROM CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS WITH HARSHAW/FILTROL CATALYST

RUN NO. COAL TESTED	9001 PRINCE HB-9	9002 DONKIN HR-9	9003 LINGAN HR-9	9004 ILLINOIS NO. 6	9005 DONKIN HR-9
Temperatures: °C					
First-Stage Reactor	400	400	400	400	420
Second-Stage Reactor	425	425	425	425	425
Back Pressure, MPa	17.24	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.006	1.047	1.044	1.028	1.058
Hydrogen Treatrate, Wt. % Dry Coal	19.3	20.0	20.2	20.5	19.8
Catalyst Age, kg coal/kg catalyst					
First-Stage: Harshaw/Filtrol 2	136	168	201	234	265
Second-Stage: Amocat 1C	40	49	59	69	78
CONVERSIONS, WT. % OF maf COAL					
Pentane Solubles plus Gases	69.2	62.8	52.6	77.0	64.8
Toluene Solubles plus Gases	84.8	82.4	75.0	90.3	84.5
THF Solubles plus Gases	92.0	92.5	90.3	95.3	93.3
PRODUCT DISTRIBUTION, WT. % OF maf COAL					
Gases	19.9	17.8	14.3	22.4	19.3
Oil	49.3	45.0	38.3	54.6	45.5
Asphaltenes	15.6	19.6	22.4	13.3	19.7
Preasphaltenes	7.2	10.1	15.3	5.0	8.8
H_2 CONSUMPTION, WT. % OF maf COAL	6.9	6.6	6.1	7.7	6.8

TABLE 8 (Continued)

RESULTS FROM CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS WITH HARSHAW/FILTROL CATALYST

RUN NO. COAL TESTED	9006 DONKIN HR-9	9007 DONKIN HR-9	9008 DONKIN HR-9	9009 DONKIN HR-9
Temperatures: °C				
First-Stage Reactor	420	400	400	400
Second-Stage Reactor	425	425	425	440
Back Pressure, MPa	20.68	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.047	1.035	1.012	1.031
Hydrogen Treatrate, Wt. % Dry Coal	20.0	20.2	20.7	20.3
Catalyst Age, kg coal/kg catalyst				
First-Stage: Harshaw/Filtrol 2	296	327	30	61
Second-Stage: Amocat 1C	87	96	105	114
CONVERSIONS, WT. % OF maf COAL				
Pentane Solubles plus Gases	66.5	57.0	60.2	65.5
Toluene Solubles plus Gases	85.5	80.3	80.9	85.9
THF Solubles plus Gases	93.6	93.0	93.2	93.4
PRODUCT DISTRIBUTION, WT. % OF maf COAL				
Gases	19.1	18.1	17.9	19.2
Oil	47.4	38.9	42.3	46.3
Asphaltenes	19.0	23.3	20.7	20.4
Preasphaltenes	8.1	12.7	12.3	7.5
H_2 CONSUMPTION, WT. % OF maf COAL	6.7	6.5	7.1	7.2

5.1.1 Coal Reactivity

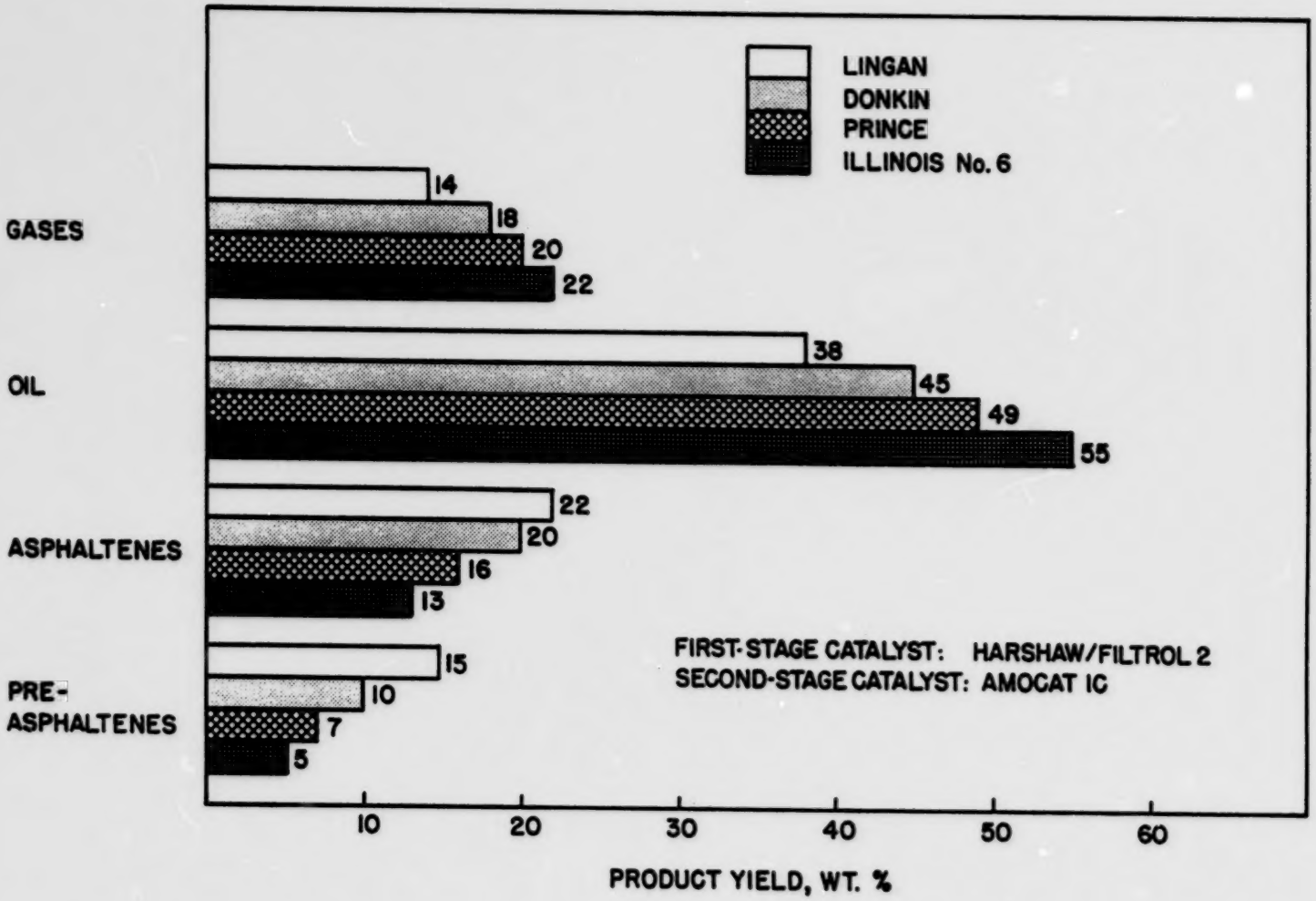
The three Sydney area coals and Illinois No. 6 were tested at similar operating conditions in Run Nos. 9001 - 9004 inclusive. The first and second-stage reactor temperatures were 400 and 425°C respectively. Slurry feedrate was targeted at 1.0 kg/hr and system pressure was maintained at 17.24 MPa. High coal conversion was achieved in all four experiments, 90 - 95%.

Product distribution varied considerably as a function of coal feedstock as illustrated in Figure 3. Gas yields ranged from 14% with Lingan Harbour seam coal to 22% with Illinois No. 6. Prince Hub, Donkin Harbour and Lingan Harbour seam coals produced 49, 45 and 38% oil respectively compared with 55% from Illinois No. 6. Lingan produced the highest yields of asphaltenes and preasphaltenes at 22 and 15%. The three Sydney area coals yielded higher levels of asphaltenes and preasphaltenes than the American coal. Hydrogen consumption ranged from 6.1% in Run No. 9003 with Lingan coal to 7.7% in Run No. 9004 with Illinois No. 6.

The order of coal reactivity established is as follows: Illinois No. 6 > Prince Hub > Donkin Harbour > Lingan Harbour. This order is quite similar to the trend in activity observed in earlier studies. The difference in coal reactivity is primarily attributed to coal rank. Simple correlations show the dependence of coal reactivity on rank, defined in terms of maximum reflectance of vitrinite, (% R_o Max.)

and composition in terms of reflectance types. For example, the most reactive coal, Illinois No. 6, has the lowest vitrinite reflectance, 0.60, and the lowest reflectance types, V-5 and V-6.

Figure 3 – PRODUCT DISTRIBUTION AS A FUNCTION OF COAL FEEDSTOCK USING HARSHAW/FILTROL 2



5.1.2 Effect of Total Pressure

Run Nos. 9005 and 9006 show the effect of operating pressure on process performance using Donkin coal. The former test was conducted at 17.24 MPa, the latter at 20.68 MPa. Coal conversion in both runs was quite similar, ca. 93%. Gas production was essentially the same at 19%. Oil yield was slightly higher in Run No. 9006 at 20.68 MPa, 47.4%, compared with 45.5% in Run No. 9005. Measured hydrogen consumption was essentially the same at both pressures investigated.

5.1.3 Interaction of Temperature and Catalyst Ageing

An examination of the data from Run Nos. 9002, 9005 and 9007 illustrates the influence of reaction temperature and catalyst ageing on raw product yields. A comparison of Run No. 9002 carried out at a first-stage reactor temperature of 400°C with Run No. 9005 conducted at 420°C shows that both runs produced very similar coal conversion and oil yield. The former run produced 92.5% coal conversion with 45.0% pentane soluble oil whereas the latter produced 93.3% coal conversion with 45.5% pentane soluble oil. The higher first-stage reactor temperature in Run No. 9005 was expected to produce more oil than it actually did.

A comparison of product distribution in Run Nos. 9002 and 9007 indicates that catalyst deterioration occurred quite rapidly in the system. Oil yield decreased from 45.0% in Run No. 9002 to 38.9% in Run No. 9007. Overall process performance declined substantially as catalyst in the first and second-stage reactor aged from 168 and 49 kg coal/kg

catalyst respectively to 327 and 96 kg coal/kg catalyst. These results provide an explanation for the lower-than-expected oil yield in Run No. 9005. Apparently, the degree of catalyst fouling off-set the positive influence of higher temperature observed in earlier work as the first-stage reactor temperature was raised from 400 to 420°C.

After Run No. 9007 the first-stage reactor was opened to change-out the catalyst. Two coal liquefaction experiments were conducted with the fresh Harshaw/Filtrol catalyst to further investigate its performance. The fresh catalyst was sulphided prior to the liquefaction tests. Run No. 9008 was made at operating conditions similar to Run No. 9002. Although hydrogen consumption was higher in Run No. 9008, the yield of pentane soluble oil was only 42.3% compared with 45.0% in Run No. 9002.

Assuming that the activity of the first charge of Harshaw/Filtrol catalyst declined quite rapidly with age, it follows that the quality of the first-stage reactor effluent also deteriorated as a function of time. Thus, the level of asphaltenes and preasphaltenes in the second-stage reactor feed increased as the test program progressed. Increasing the feed of heavy asphaltenic, hydrogen-lean components to the second-stage catalytic reactor would cause more rapid fouling of the catalyst. With this in mind, the second-stage reactor temperature was raised by 15° to 440°C in Run No. 9009 to compensate for decreased catalytic activity. This run produced the highest oil yield from Donkin coal, 46.3%.

5.2 Performance Comparison of First-Stage Nickel-Molybdenum Bimodal Catalysts

The evaluation of hydroprocessing catalysts is an essential part of our program on advanced two-stage coal liquefaction. Recently, experimental work has been concentrated on catalytic two-stage processing with emphasis on first-stage catalyst performance. Two nickel-molybdenum bimodal catalysts have been tested in the first-stage reactor thus far, Amocat 1C and Harshaw/Filtrol 2. The catalysts differ primarily in metals loading, modal pore size distribution and surface area as shown in Table 4. The Amocat catalyst has lower metals content, smaller mean mesopore and macropore diameters and higher surface area than the Harshaw/Filtrol material.

Coal conversion and raw product distribution were used as parameters in evaluating the performance of the catalysts. Direct comparisons were made between runs at similar operating conditions. Amocat 1C was used as the second-stage reactor catalyst in all of the experiments. Details pertaining to the runs are given in the previous section and an earlier report (10).

High coal conversion, 90 - 95%, was achieved with both catalysts over the range of operating conditions investigated, see Table 9. Product yields are illustrated for Lingan Harbour, Donkin Harbour, Prince Hub and Illinois No. 6 coals in Figure 4. In the experiments with the three Sydney area coals, Amocat 1C provided higher

coal-to-oil conversion activity than the Harshaw/Filtrol catalyst at similar operating conditions including catalyst age. In runs with Illinois No. 6 coal, the higher aged Amocat 1C catalyst produced more oil and less asphaltene/preasphaltene components than the less used Harshaw/Filtrol catalyst. These results show that Amocat 1C is superior to Harshaw/Filtrol 2 in terms of activity and stability.

In addition, these data suggest that coal-to-oil conversion selectivity responds primarily to catalyst activity. The superior performance of Amocat 1C is mainly attributed to its high surface area in combination with suitable type and adequate amount of catalytic metals. Furthermore, it seems that the range of pore sizes in this catalyst does not present serious reactant diffusion limitations. In fact, the pore sizes and pore size distribution may contribute to the higher stability of this catalyst. The smaller pores may exclude heavy hydrocarbon components, coke precursors, thereby limiting coke fouling on interior active surfaces.

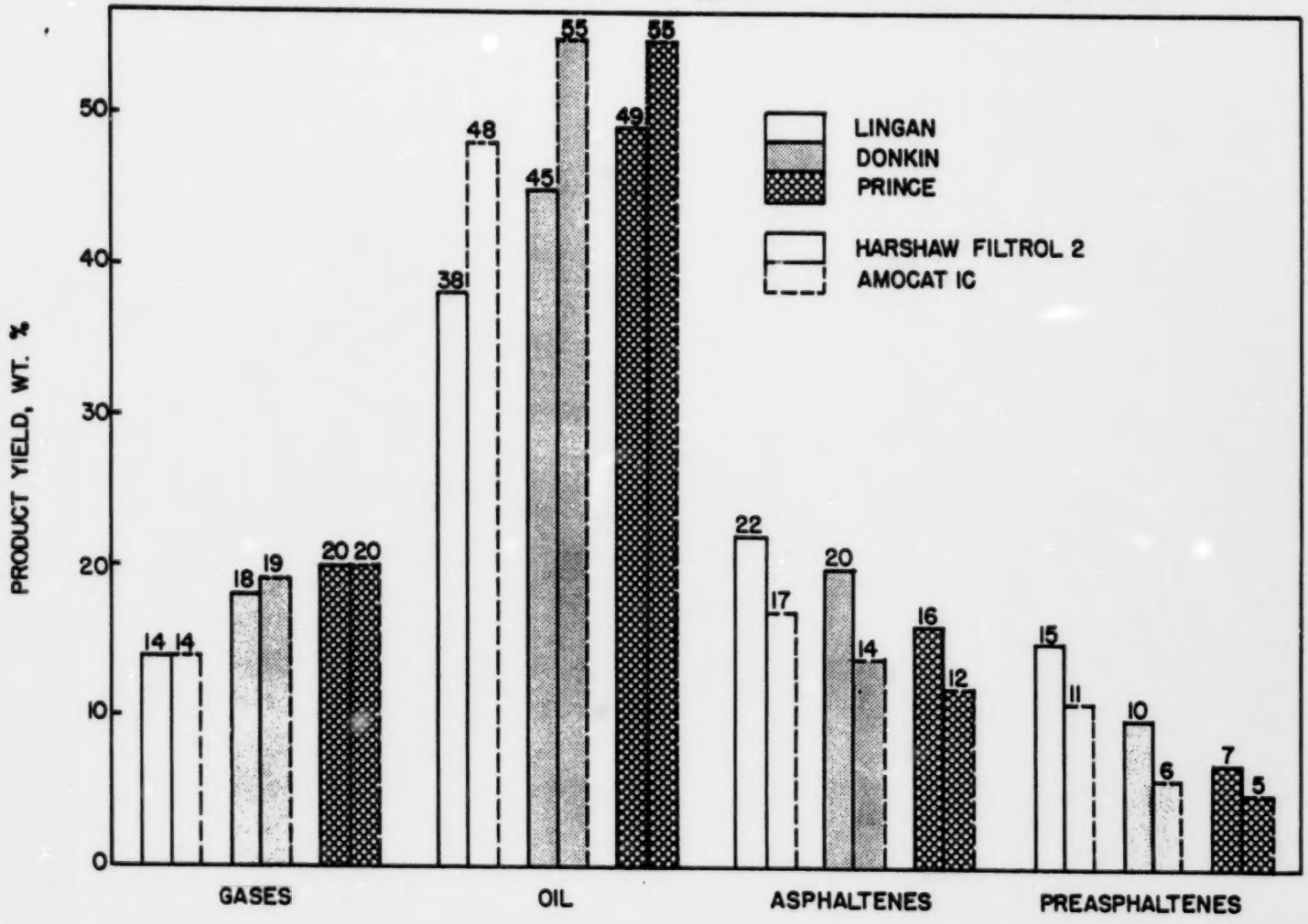
Coal liquefaction places severe demands on hydroprocessing catalysts. In order to achieve high distillate yields and product quality, supported catalyst will require relatively large surface area, well controlled pore size distribution and compositions which are resistant to coke fouling. Unfortunately, catalyst deactivation by coke deposition and metals fouling is still a serious problem. Improvements are required to extend catalyst life at the high severity operating conditions of this application.

TABLE 9

COMPARISON OF CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS WITH HARSHAW/FILTROL 2 AND AMOCAT 1C CATALYSTS

RUN NO. COAL TESTED FIRST-STAGE CATALYST	9001 PRINCE HB-9 H/F 2	8003 PRINCE HB-8 AMOCAT 1C	9002 DONKIN HR-9 H/F 2	8004 DONKIN HR-8 AMOCAT 1C	9003 LINGAN HR-9 H/F 2	8005 LINGAN HR-8 AMOCAT 1C	9004 ILLINOIS NO. 6 H/F 2	8011 ILLINOIS NO. 6 AMOCAT 1C
Temperatures: °C								
First-Stage Reactor	400	400	400	400	400	400	400	400
Second-Stage Reactor	425	425	425	425	425	425	425	425
Back Pressure, MPa	17.24	17.24	17.24	17.24	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.006	1.006	1.047	1.041	1.044	1.048	1.028	1.040
Hydrogen Treatrate, Wt. % Dry Coal	19.3	19.8	20.0	19.3	20.2	19.1	20.5	19.3
Catalyst Age, kg coal/kg catalyst								
First-Stage Reactor	136	144	168	175	201	206	234	392
Second-Stage Reactor	40	42	49	51	59	60	69	114
CONVERSIONS, WT. % OF maf COAL								
Pentane Solubles plus Gases	69.2	75.0	62.8	73.5	52.6	61.8	77.0	84.1
Toluene Solubles plus Gases	84.8	87.3	82.4	87.2	75.0	78.9	90.3	91.3
THF Solubles plus Gases	92.0	92.2	92.5	92.9	90.3	90.0	95.3	94.3
PRODUCT DISTRIBUTION, WT. % OF maf COAL								
Gases	19.9	19.6	17.8	18.8	14.3	14.1	22.4	25.0
Oil	49.3	55.4	45.0	54.7	38.3	47.7	54.6	59.1
Asphaltenes	15.6	12.3	19.6	13.7	22.4	17.1	13.3	7.2
Preasphaltenes	7.2	4.9	10.1	5.7	15.3	11.1	5.0	3.0
H_2 CONSUMPTION, WT. % OF maf COAL	6.9	8.3	6.6	7.6	6.1	6.6	7.7	9.1

Figure 4 – PRODUCT DISTRIBUTION FOR SYDNEY-AREA COALS AS A FUNCTION OF FIRST-STAGE CATALYST



5.3 Operations at Reduced Coal Space Velocity

On completion of test work with the Harshaw/Filtrol catalyst the two-stage reaction zone of our coal liquefaction unit was modified. The new 3.8-liter reactor described in Task 2 was installed in place of the existing 1.5-liter first-stage reactor. The purpose of this was to investigate process performance at reduced coal space velocity in the first-stage. The overflow pipe in the new reactor was adjusted to provide one-half the slurry space velocity used in previous experiments.

Amocat 1C catalyst was used in both reactors in this study. The first and second-stage reactors were charged with 0.2 and 0.3 kg of catalyst respectively. Prior to initiating coal liquefaction experiments a 30-hour run was carried out to sulphide the fresh catalyst using hydrogenated anthracene oil spiked with dimethyl disulphide. The catalyst sulphiding procedure was followed immediately by a 24-hour run with Lingan coal to condition the catalyst. After this a series of ten experiments were conducted varying first and second-stage reactor temperatures and coal feedstock to determine raw product yields and assess catalyst stability.

A summary of the operating conditions and results for these experiments is shown in Table 10. Detailed performance and material balance data is given in Appendices B and C.

TABLE 10

CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS AT LOW FIRST-STAGE SLURRY SPACE VELOCITY

RUN NO. COAL TESTED	9010 LINGAN HR-9	9011 DONKIN HR-9	9012 DONKIN HR-9	9014 DONKIN HR-9	9015 DONKIN HR-9	9016 ILLINOIS NO. 6
Temperatures: °C						
First-Stage Reactor	415	400	420	440	420	400
Second-Stage Reactor	425	425	425	425	440	425
Back Pressure, MPa	17.24	17.24	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.536	1.025	1.037	1.020	1.008	1.038
Hydrogen Treatrate, Wt. % Dry Coal	14.6	20.5	20.3	20.7	20.9	20.3
Catalyst Age, kg coal/kg catalyst						
First-Stage: Amocat 1C	47	60	73	86	99	112
Second-Stage: Amocat 1C	31	40	49	58	67	76
CONVERSIONS, WT. % OF maf COAL						
Pentane Solubles plus Gases	62.8	78.2	81.4	85.7	83.0	84.3
Toluene Solubles plus Gases	82.3	89.8	91.2	93.2	91.6	92.2
THF Solubles plus Gases	91.4	93.9	93.9	94.8	94.4	95.3
PRODUCT DISTRIBUTION, WT. % OF maf COAL						
Gases	14.2	20.2	22.3	26.7	25.9	23.8
Oil	48.6	58.0	59.1	59.0	57.1	60.5
Asphaltenes	19.5	11.6	9.8	7.5	8.6	7.9
Preasphaltenes	9.1	4.1	2.7	1.6	2.8	3.1
H_2 CONSUMPTION, WT. % OF maf COAL	7.1	8.3	9.7	10.5	9.8	9.9

TABLE 10 (Continued)

CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS AT
LOW FIRST-STAGE SLURRY SPACE VELOCITY

RUN NO. COAL TESTED	9017 LINGAN HR-9	9018 PRINCE HB-9	9019 DONKIN HR-9	9020 LINGAN HR-9	9022A ILLINOIS NO.6	9022B ILLINOIS NO. 6
Temperatures: °C						
First-Stage Reactor	440	420	420	420	400	420
Second-Stage Reactor	425	425	425	425	425	425
Back Pressure, MPa	17.24	17.24	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.019	1.043	1.024	1.033	1.054	1.034
Hydrogen Treatrate, Wt. % Dry Coal	20.7	20.1	20.6	20.4	20.1	20.4
Catalyst Age, kg coal/kg catalyst						
First-Stage: Amocat 1C	125	138	151	164	191	197
Second-Stage: Amocat 1C	85	94	103	112	131	135
CONVERSIONS, WT. % OF maf COAL						
Pentane Solubles plus Gases	77.0	77.7	73.0	65.8	77.5	84.9
Toluene Solubles plus Gases	89.0	88.9	87.5	83.5	89.7	92.9
THF Solubles plus Gases	93.6	93.1	94.0	92.6	95.2	95.9
PRODUCT DISTRIBUTION, WT. % OF maf COAL						
Gases	23.2	23.2	21.8	18.4	22.3	25.6
Oil	53.8	54.5	51.2	47.4	55.2	59.3
Asphaltenes	12.0	11.2	14.5	17.7	12.2	8.0
Preasphaltenes	4.6	4.2	6.5	9.1	5.5	3.0
H_2 CONSUMPTION, WT. % OF maf COAL	9.7	8.1	8.5	8.0	8.9	10.1

5.3.1 Influence of First-Stage Reactor Temperature

Figure 5 illustrates the influence of first-stage reactor temperature on conversion and raw product yields using Donkin Harbour seam coal. Run Nos. 9011, 9012 and 9014 were carried out at first-stage temperatures of 400, 420 and 440°C respectively with the second-stage reactor held at 425°C as the first-stage temperature was raised from 400 to 440°C; coal conversion remained essentially the same at 94-95%, gas production increased from 20.2 to 26.7%, oil yield remained fairly constant around 59%, asphaltenes decreased from 11.6 to 7.5% and preasphaltenes decreased from 4.1 to 1.6%. Measured hydrogen consumption rose from 8.3% at 400°C in Run No. 9011 to 10.5% at 440°C in Run No. 9014.

Run No. 9017 was made with Lingan Harbour seam coal at a first-stage temperature of 440°C. This run produced the highest oil yield, 53.8%, and the highest hydrogen consumption, 9.7%, observed with this coal to date. Asphaltene and preasphaltene levels were substantially reduced by the combination of high temperature and low coal space velocity in the first-stage reactor.

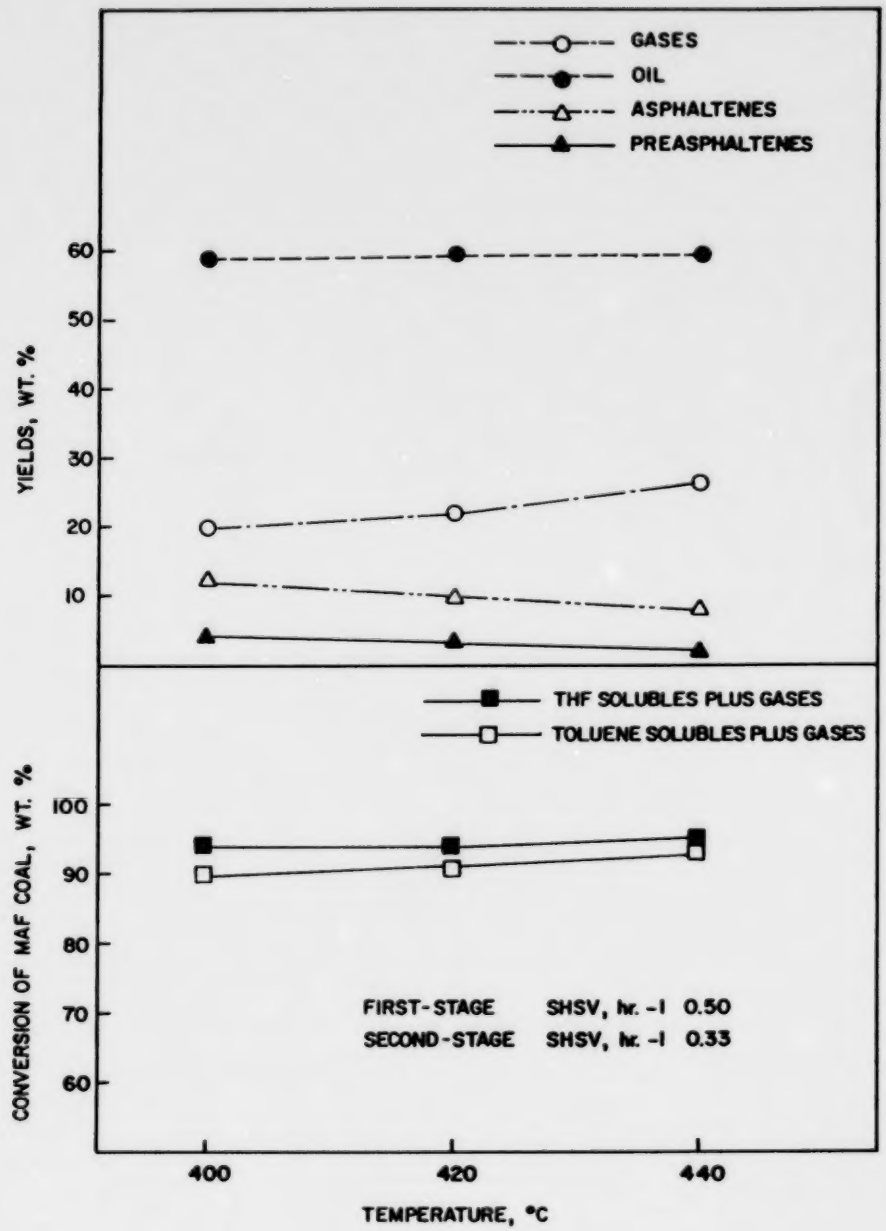


Figure 5 - EFFECT OF FIRST-STAGE TEMPERATURE ON COAL CONVERSION AT LOW SHSV

5.3.2 Effect of Second-Stage Reactor Temperature

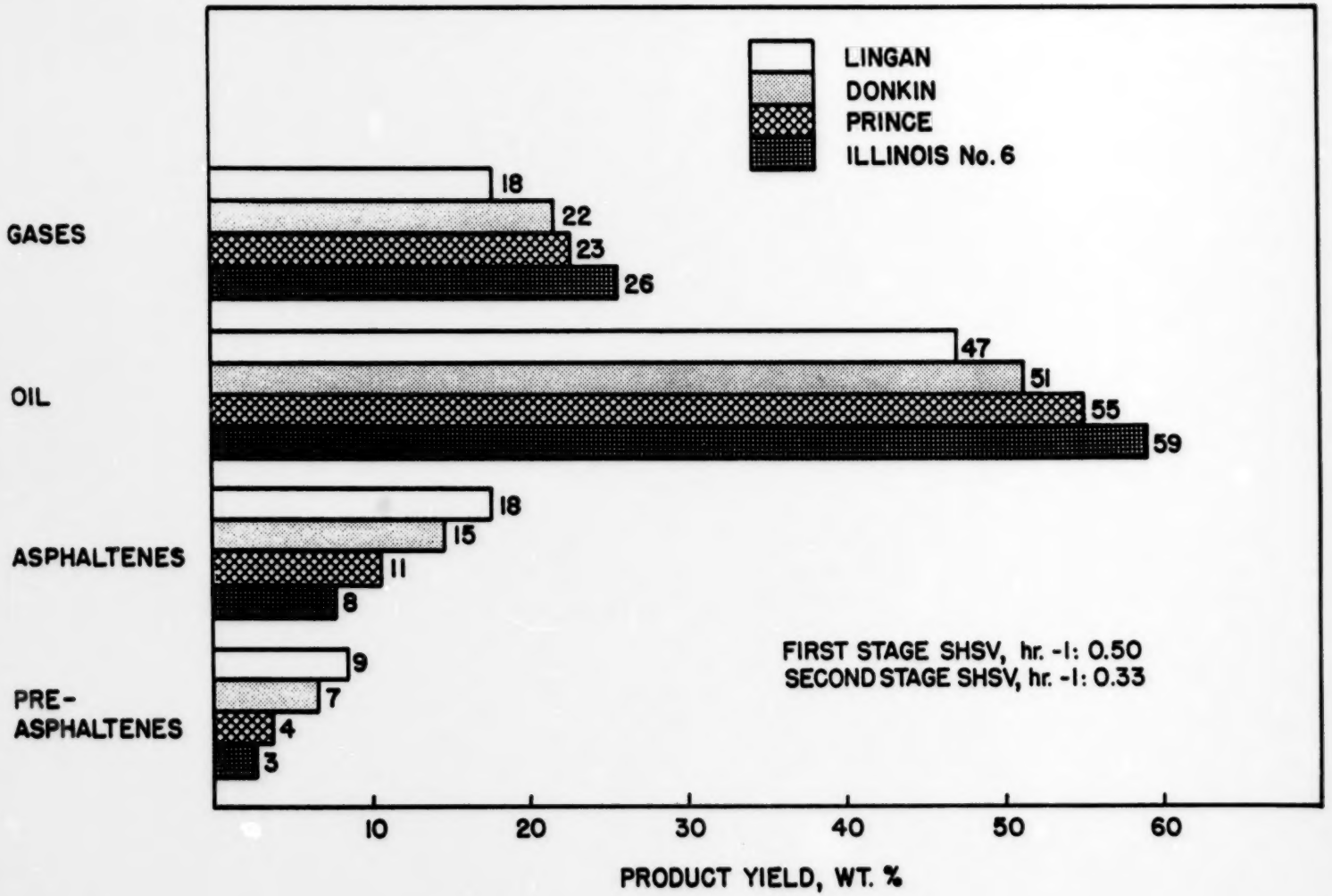
The second-stage reactor temperature was raised from 425°C in Run No. 9012 to 440°C in Run No. 9015 with the first-stage reactor held at 420°C. The increase of 15°C in the second-stage temperature affected a marginal increase in gas production accompanied by a marginal decrease in oil yield and asphaltenic components. Gas production and oil yield in Run No. 9015 were 25.9 and 57.1% respectively compared with 22.3 and 59.1% in Run No. 9012. Hydrogen consumption was essentially the same in both runs at about 9.8%.

5.3.3 Product Yield as a Function of Coal Feedstock

Run Nos. 9018, 9019, 9020 and 9022B were conducted using Prince Hub, Donkin Harbour, Lingan Harbour and Illinois No. 6 coals at similar operating conditions. The first and second-stage temperatures were 420 and 425°C respectively. Slurry feedrate was 1.0 kg/hr at 17.24 MPa total pressure. High coal conversion, 93-96%, was achieved in all four experiments.

Figure 6 shows the variation in raw product yields as a function of coal feedstock. Gas production ranged from 18.4% with Lingan Harbour seam coal to 25.6% with Illinois No. 6. Oil yields from Lingan Harbour, Donkin Harbour and Prince Hub were 47, 51 and 55% compared with 59% from Illinois No. 6. As expected, the three Sydney area coals produced higher levels of asphaltenes and preasphaltenes than the lower rank American coal. In fact, the Harbour seam coals yielded 2 to 2.5 times more of these components than Illinois No. 6.

Figure 6 – PRODUCT DISTRIBUTION AS A FUNCTION OF COAL FEEDSTOCK AT LOW SHSV



5.3.4 Influence of Catalyst Age

Data presented in Table 11 shows a decrease in catalyst activity with age.

Run Nos. 9012 and 9019 were carried out using Donkin Harbour coal. First and second-stage temperatures were 420 and 425°C respectively. A comparison of results shows that process performance declined as catalyst in the first and second-stage reactors aged from 73 and 49 kg coal/kg catalyst respectively in Run No. 9012 to 151 and 103 kg coal/kg catalyst in Run No. 9019. Oil yield decreased from 59.1% in the former run to 51.2% in the latter while asphaltene/preasphaltene components increased from 12.5 to 21.0% and hydrogen consumption declined from 9.7 to 8.5%.

Similar trends were observed in Run Nos. 9016 and 9022A with Illinois No. 6 coal at first and second-stage temperatures of 400 and 425°C. As the catalyst in the first and second-stage reactors aged from 112 and 76 kg coal/kg catalyst respectively in Run No. 9016 to 191 and 131 kg coal/kg catalyst in Run No. 9022A, oil yield decreased from 60.5% to 55.2% while hydrogen consumption declined from 9.9 to 8.9%.

Elemental analysis of pentane, toluene and THF soluble fractions from all experiments are shown in Table 12. Comparisons of Run No. 9012 with Run No. 9019 and Run No. 9016 with Run No. 9022A show that the levels of nitrogen and sulphur in these fractions increased as the catalyst aged.

TABLE 11

RAW PRODUCT DISTRIBUTION AS A FUNCTION OF CATALYST AGE

RUN NO. COAL TESTED	9012 DONKIN HR-9	9019 DONKIN HR-9	9016 ILLINOIS NO. 6	9022A ILLINOIS NO. 6
Temperatures, °C:				
First-Stage Reactor	420	420	400	400
Second-Stage Reactor	425	425	425	425
Back Pressure, MPa	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.037	1.024	1.038	1.054
Hydrogen Treatrate, Wt. % Dry Coal	20.3	20.6	20.3	20.1
Catalyst Age, kg coal/kg catalyst				
First-Stage: Amocat 1C	73	151	112	191
Second Stage: Amocat 1C	49	103	76	131
CONVERSIONS, WT. % OF maf COAL				
Pentane Solubles plus Gases	81.4	73.0	84.3	77.5
Toluene Solubles plus Gases	91.2	87.5	92.2	89.7
THF Solubles plus Gases	93.9	94.0	95.3	95.2
PRODUCT DISTRIBUTION, WT. % OF maf COAL				
Gases	22.3	21.8	23.8	22.3
Oil	59.1	51.2	60.5	55.2
Asphaltenes	9.8	14.5	7.9	12.2
Preasphaltenes	2.7	6.5	3.1	5.5
H_2 CONSUMPTION, WT. % OF maf COAL	9.7	8.5	9.9	8.9

TABLE 12

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9000	9001	9002	9003	9004	9005	9006	9007
Pentane Soluble Fraction								
Carbon, Wt. %	89.81	89.98	90.21	89.64	89.56	89.37	89.69	89.48
Hydrogen, Wt. %	9.46	9.45	9.33	9.27	9.37	9.24	9.41	9.33
Nitrogen, Wt. %	0.22	0.15	0.17	0.16	0.15	0.17	0.14	0.19
Sulphur, Wt. %	0.021	0.035	0.031	0.026	0.025	0.028	0.032	0.039
Toluene Soluble Fraction								
Carbon, Wt. %	89.70	89.03	89.98	90.83	89.72	89.25	89.84	89.30
Hydrogen Wt. %	9.27	9.26	9.21	9.12	9.30	9.14	9.36	8.98
Nitrogen, Wt. %	0.32	0.21	0.25	0.30	0.22	0.25	0.27	0.29
Sulphur, Wt. %	0.026	0.060	0.057	0.041	0.051	0.057	0.064	0.074
THF Soluble Fraction								
Carbon, Wt. %	89.63	87.75	89.73	89.51	89.40	89.01	89.25	89.17
Hydrogen, Wt. %	8.93	9.16	9.14	9.05	9.01	9.03	9.24	8.93
Nitrogen, Wt. %	0.37	0.26	0.31	0.39	0.29	0.29	0.26	0.35
Sulphur, Wt. %	0.043	0.072	0.089	0.055	0.086	0.083	0.088	0.110

TABLE 12 (Continued)

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9008	9009	9010	9011	9012	9014	9015	9016
Pentane Soluble Fraction								
Carbon, Wt. %	90.40	89.84	89.79	89.86	90.13	90.01	90.03	90.05
Hydrogen, Wt. %	9.60	9.11	9.25	9.50	9.34	9.47	9.30	9.51
Nitrogen, Wt. %	0.18	0.18	0.18	0.15	0.13	0.12	0.12	0.14
Sulphur, Wt. %	0.034	0.034	0.017	0.023	0.024	0.025	0.019	0.023
Toluene Soluble Fraction								
Carbon, Wt. %	90.12	89.07	89.84	90.09	89.85	89.45	89.69	89.83
Hydrogen, Wt. %	9.22	8.76	9.11	9.22	9.14	9.21	8.94	9.25
Nitrogen, Wt. %	0.27	0.28	0.25	0.18	0.16	0.16	0.16	0.17
Sulphur, Wt. %	0.060	0.068	0.026	0.035	0.037	0.035	0.030	0.047
THF Soluble Fraction								
Carbon, Wt. %	89.76	88.72	89.65	89.83	90.14	90.17	89.94	89.97
Hydrogen, Wt. %	9.12	8.79	8.77	9.10	9.05	9.05	8.82	9.08
Nitrogen, Wt. %	0.34	0.33	0.35	0.20	0.18	0.18	0.19	0.20
Sulphur, Wt. %	0.083	0.078	0.034	0.054	0.047	0.046	0.046	0.067

TABLE 12 (Continued)

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9017	9018	9019	9020	9022A	9022B
Pentane Soluble Fraction						
Carbon, Wt. %	89.83	89.77	90.07	89.78	90.09	89.93
Hydrogen, Wt. %	9.29	9.33	9.53	9.10	9.48	9.47
Nitrogen, Wt. %	0.15	0.14	0.13	0.15	0.15	0.14
Sulphur, Wt. %	0.015	0.020	0.029	0.017	0.027	0.027
Toluene Soluble Fraction						
Carbon, Wt. %	89.56	90.00	89.67	89.65	89.35	89.81
Hydrogen, Wt. %	9.09	9.11	9.08	8.90	9.22	9.39
Nitrogen, Wt. %	0.20	0.18	0.19	0.24	0.21	0.19
Sulphur, Wt. %	0.022	0.040	0.050	0.032	0.054	0.044
THF Soluble Fraction						
Carbon, Wt. %	89.94	89.36	89.41	89.81	89.43	89.52
Hydrogen, Wt. %	9.13	9.15	9.16	8.92	9.22	9.32
Nitrogen, Wt. %	0.22	0.21	0.23	0.28	0.26	0.21
Sulphur, Wt. %	0.039	0.060	0.078	0.046	0.084	0.070

5.4 Comparison of Process Performance at Low and Medium First-Stage Slurry Space Velocity

Experimental results are presented in Table 13 to assess process performance as a function of slurry space velocity in the first-stage reactor. Process performance being defined in terms of coal conversion, product distribution and hydrogen consumption. The 9000 series experiments are at low slurry space velocity and hence low coal space velocity whereas the 8000 series runs are at medium space velocity. The 8000 series runs were done during the first phase of work on catalytic two-stage liquefaction (10).

Run Nos. 9010 and 8002 were both catalyst conditioning runs conducted with Lingan Harbour coal. First and second-stage temperatures were 415 and 425°C respectively. Slurry feedrate was approximately 1.5 kg/hr at 17.24 MPa total pressure. In Run No. 9010 the first-stage slurry hourly space velocity (SHSV) was 0.75 hr^{-1} compared with 1.50 hr^{-1} in Run No. 8002. The second-stage SHSV was 0.50 hr^{-1} in both experiments. Run No. 9010 produced more oil and less asphaltene/preasphaltene components than Run No. 8002. Oil yield was 48.6% in Run No. 9010 compared with 44.1% in Run No. 8002. The former run also exhibited higher hydrogen consumption than the latter, 7.1% compared to 5.2%.

Run Nos. 9011 and 8004 were done with Donkin Harbour coal. The first and second-stage temperatures were 400 and 425°C respectively. Slurry feedrate was 1.0 kg/hr at 17.24 MPa total pressure. The first-

stage SHSV was 0.50 hr^{-1} in Run No. 9011 compared with 1.00 hr^{-1} in Run No. 8004. The second-stage SHSV was 0.33 hr^{-1} in both tests. Run No. 9011 exhibited higher oil yield, lower asphaltene/preasphaltene levels and higher hydrogen consumption than Run No. 8004. These trends were similar to the ones observed in Run Nos. 9010 and 8002.

Run Nos. 9018 and 8009 were carried out with Prince Hub coal at first and second-stage temperatures of 420 and 425°C respectively. Slurry feedrate was 1.0 kg/hr at 17.24 MPa total pressure. The first-stage SHSV was 0.50 hr^{-1} in the former run compared with 1.00 hr^{-1} in the latter. The second-stage SHSV was 0.33 hr^{-1} in both experiments. Run No. 9018 yielded slightly less oil at lower measured hydrogen consumption. Furthermore, asphaltene/preasphaltene levels were essentially the same in Run No. 9018 and Run No. 8009. The apparent decline in process performance at reduced first-stage slurry space velocity may be attributed to catalyst fouling.

The results presented herein demonstrate improved process performance at reduced first-stage coal space velocity with fresh, highly-active catalyst. However, the results suggest that longer residence time in the first-stage reactor increases the rate of catalyst fouling. It should be noted at this point that the first-stage catalyst in the 9000 series test work was used at higher temperature, 440°C, before Run No. 9018 than it was in the 8000 series prior to Run No. 8009. This high temperature service may have contributed to catalyst deactivation leading to the observed decline in process performance.

TABLE 13

CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS AT LOW AND MEDIUM SHSV

RUN NO. COAL TESTED	9010 LINGAN HR-9	8002 LINGAN HR-6	9011 DONKIN HR-9	8004 DONKIN HR-8	9018 PRINCE HB-9	8009 PRINCE HB-8
Temperatures, °C:						
First-Stage Reactor	415	415	400	400	420	420
Second-Stage Reactor	425	425	425	425	425	425
Back Pressure, MPa	17.24	17.24	17.24	17.24	17.24	17.24
Slurry Feedrate, kg/hr	1.536	1.546	1.025	1.041	1.043	1.000
Hydrogen Treatrate, Wt. % Dry Coal	14.6	9.7	20.5	19.3	20.1	20.3
Catalyst Age, kg coal/kg catalyst						
First-Stage: Amocat 1C	47	107	60	175	138	330
Second-Stage: Amocat 1C	31	31	40	51	94	96
SHSV, hr^{-1}						
First-Stage Reactor	0.75	1.50	0.50	1.00	0.50	1.00
Second-Stage Reactor	0.50	0.50	0.33	0.33	0.33	0.33
CONVERSIONS, WT. % OF maf COAL						
Pentane Solubles plus Gases	62.8	57.1	78.2	73.5	77.7	77.2
Toluene Solubles plus Gases	82.3	77.8	89.8	87.2	88.9	87.8
THF Solubles plus Gases	91.4	90.5	93.9	92.9	93.1	92.3
PRODUCT DISTRIBUTION, WT. % OF maf COAL						
Gases	14.2	13.0	20.2	18.8	23.2	21.1
Oil	48.6	44.1	58.0	54.7	54.5	56.1
Asphaltenes	19.5	20.7	11.6	13.7	11.2	10.6
Preasphaltenes	9.1	12.7	4.1	5.7	4.2	4.5
H_2 CONSUMPTION, WT. % OF maf COAL	7.1	5.2	8.3	7.6	8.1	8.8

CONCLUSIONS AND RECOMMENDATIONS

The second part of a research program on catalytic two-stage liquefaction of Nova Scotia bituminous coals has been completed. Bench-scale studies were focused on catalyst performance and coal space velocity in the first-stage reaction zone. Experiments were conducted in a once-through mode using hydrogenated coal-derived oil as reaction solvent. Harbour and Hub seam coals from the Sydney coalfield were tested along with Illinois No. 6 coal for comparative purposes.

Studies on catalytic two-stage liquefaction were initiated using Amocat 1C nickel-molybdenum bimodal catalyst in both reactors. On completion of that work it was decided to test a first-stage catalyst with higher metals loading and larger mean mesopore and macropore diameters than Amocat 1C. A total of ten experiments were carried out with the selected catalyst, Harshaw/Filtrol 2. Amocat 1C was used as the second-stage catalyst in these experiments. The runs were made to investigate coal reactivity and the effects of reaction temperature, total pressure and catalyst age on conversion and product yield.

High coal conversions, 90-95%, were achieved with all four coals at a first-stage reactor temperature of 400°C. However, product distribution varied widely as a function of coal feedstock. Gas production ranged from 14% with Lingan Harbour seam coal to 22% with Illinois No. 6. Lingan Harbour, Donkin Harbour and Prince Hub coals

produced 38, 45 and 49% oil compared with 55% from Illinois No. 6. The three Sydney area coals produced substantially higher levels of asphaltenes and preasphaltenes than the lower rank American coal. Lingan produced the highest yields of asphaltenes and preasphaltenes at 22 and 15% respectively. The order of coal reactivity established is as follows: Illinois No. 6 > Prince Hub > Donkin Harbour > Lingan Harbour.

Experiments with Donkin coal showed that increasing total pressure from 17.24 to 20.68 MPa had little effect on conversion and raw product distribution. Coal conversion was constant at 93%. Oil yield was slightly higher at 20.68 MPa, 47% as compared with 45% at 17.24 MPa. Catalyst age, however, had a significant effect on product distribution. As the first and second-stage catalysts aged from 168 and 49 kg coal/kg catalyst respectively to 327 and 96 kg coal/kg catalyst, oil yield decreased from 45 to 39%. The decrease in oil yield was accompanied by an increase in asphaltene/preasphaltene components from 30 to 36%.

A comparison of experimental results shows that Amocat 1C is superior to Harshaw/Filtrol 2 in terms of activity and stability. The data suggest that coal-to-oil conversion selectivity responds primarily to catalyst activity. The superior performance of Amocat 1C is mainly attributed to its high surface area in combination with suitable type and adequate amount of catalytic metals. Furthermore, it seems that the range of pore sizes in this catalyst does not present serious reactant diffusion limitations.

The catalyst performance evaluation was followed by a series of experiments at reduced coal space velocity in the first-stage. A new 3.8-liter reactor was installed in place of the existing 1.5-liter first-stage reactor to provide one-half the slurry space velocity used in earlier tests. Amocat 1C catalyst was used in both first and second-stage reactors. Coal conversion and raw product yield were established as a function of reaction temperature, coal feedstock and catalyst age.

In experiments with Donkin Harbour coal, as the first-stage temperature was raised from 400 to 440°C; coal conversion was fairly constant at about 94%, gas production increased from 20 to 27%, oil yield was fairly constant at 59%, asphaltenes decreased from 12 to 8% and preasphaltenes decreased from 4 to 2%.

High coal conversions, 93-94%, were achieved with all three Sydney area coals as expected. Product yields varied considerably as in previous coal feedstock investigations. In experiments at similar operating conditions and catalyst age, gas production ranged from 18% with Lingan coal to 23% with Prince. Oil yields from Lingan Harbour, Donkin Harbour and Prince Hub were 47, 51 and 55% respectively. Lingan coal produced the highest level of asphaltene/preasphaltene components, 27%.

Process performance was found to decline as the catalyst aged. For example, in runs with Donkin coal, oil yield decreased from 59 to 51% as the first and second-stage catalyst aged from 73 and 49 kg coal/kg

catalyst respectively to 151 and 103 kg coal/kg catalyst. Furthermore, asphaltene/preasphaltene components increased from 13 to 21% and hydrogen consumption decreased from 9.7 to 8.5%.

A comparison of experimental results at low and medium first-stage slurry space velocity shows improved process performance at low space velocity with fresh, highly-active catalyst. However, the results suggest that longer residence time increases catalyst fouling. In this study, higher first-stage temperature operations early in the low space velocity test sequence may have contributed to catalyst deactivation. Nevertheless, it appears that little advantage, if any, was gained operating at the reduced coal space velocity.

Based on the results of our work to date the following recommendations for future work are proposed:

(a) Undertake a laboratory program to investigate the asphaltene conversion capacity of several multi-functional hydroprocessing catalysts. Conduct catalyst screening tests in a 300 cc static basket catalytic reactor using coal liquefaction recycle liquid as feedstock.

(b) Initiate experimental work on catalyst regeneration to determine the extent to which used catalyst can be reactivated for recycle. Regenerate used catalysts by

controlled oxidation. Assess the relative activity of fresh, aged and regenerated catalyst for hydrogenation, denitrogenation and desulphurization functionalities.

(c) Continue the bench program on catalytic two-stage liquefaction. Conduct short-term exploratory trials with high activity asphaltene conversion catalysts and/or test regenerated hydroprocessing catalysts.

(d) Conduct advanced two-stage liquefaction tests with liquid recycle to assess process stability, yield distribution, product composition and catalyst aging characteristics.

At present, conventional coal liquefaction is not economically viable in Eastern Canada. However, technological advances are being made to improve the economics of converting coal into premium quality liquid fuels. Recent advances in two-stage liquefaction offer the potential to enhance liquid yield and improve hydrogen utilization. Further test work is required on two-stage process configurations applied to Sydney area coals to determine the extent to which these improvements can be realized.

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APPENDIX A

CONVERSION CALCULATIONS

APPENDIX A

CONVERSION CALCULATIONS FOR CONTINUOUS COAL LIQUEFACTION EXPERIMENTS

Calculations are based on operations during the steady state sampling period.

CONVERSIONS, WT. %:

$$\text{Pentane sol. + gases} = \frac{\text{(maf coal charged) - (organic pentane insol.)}}{\text{maf coal charged}} \times 100$$

$$\text{Toluene sol. + gases} = \frac{\text{(maf coal charged) - (organic toluene insol.)}}{\text{maf coal charged}} \times 100$$

$$\text{THF sol. + gases} = \frac{\text{(maf coal charged) - (organic THF insol.)}}{\text{maf coal charged}} \times 100$$

$$\text{Gases} = \frac{\text{product gases}}{\text{maf coal charged}} \times 100$$

DEFINITION OF PRODUCTS

Oil = pentane solubles

Asphaltenes = toluene solubles - pentane solubles

Preasphaltenes = THF solubles - toluene solubles

APPENDIX B

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS

OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO. COAL TESTED	9000 LINGAN HR-7	9001 PRINCE HB-9	9002 DONKIN HR-9	9003 LINGAN HR-9
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	17.24	17.24	17.24
Temperatures, °C:				
First-Stage Reactor	415	400	400	400
Second-Stage Reactor	425	425	425	425
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.568	1.066	1.047	1.044
Raw Coal, kg/hr	0.439	0.298	0.293	0.292
Hydrogen, L/hr	576.	618.	636.	648.
SHSV, hr^{-1}; First-Stage	1.50	1.00	1.00	1.00
Second-Stage	0.50	0.33	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	12.0	19.3	20.0	20.2
Catalyst Age, kg coal/kg catalyst				
First-Stage: Harshaw/Filtrol 2	109	136	168	201
Second-Stage: Amocat 1C	32	40	49	59
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	58.9	69.2	62.8	52.6
Toluene Solubles plus Gases	81.2	84.8	82.4	75.0
THF Solubles plus Gases	92.5	92.0	92.5	90.3
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.0	2.2	2.1	2.3
C_2H_6	1.6	2.0	2.1	1.9
C_3H_8	1.1	1.6	1.5	1.5
C_4H_{10}	1.8	3.3	2.5	2.7
CO	0.0	0.0	0.0	0.0
H_2S	0.7	2.0	1.5	1.1
H_2O	5.7	7.9	7.3	4.1
NH_3	0.7	0.9	0.8	0.7
Oil	45.3	49.3	45.0	38.3
Asphaltenes	22.3	15.6	19.6	22.4
Preasphaltenes	11.3	7.2	10.1	15.3
Unreacted Coal	7.5	8.0	7.5	9.7
D. H_2 CONSUMPTION, WT. % maf COAL	5.7	6.9	6.6	6.1

OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO. COAL TESTED	9004 ILLINOIS NO. 6	9005 DONKIN HR-9	9006 DONKIN HR-9	9007 DONKIN HR-9
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	17.24	20.68	17.24
Temperatures, °C:				
First-Stage Reactor	400	420	420	400
Second-Stage Reactor	425	425	425	425
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.028	1.058	1.047	1.035
Raw Coal, kg/hr	0.288	0.296	0.293	0.290
Hydrogen, L/hr	636.	636.	636.	636.
SHSV, hr^{-1}; First-Stage	1.00	1.00	1.00	1.00
Second-Stage	0.33	0.33	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	20.5	19.8	20.0	20.2
Catalyst Age, kg coal/kg catalyst				
First-Stage: Harshaw/Filtrol 2	234	265	296	327
Second-Stage: Amocat 1C	69	78	87	96
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	77.0	64.8	66.5	57.0
Toluene Solubles plus Gases	90.3	84.5	85.5	80.3
THF Solubles plus Gases	95.3	93.3	93.6	93.0
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.1	2.6	2.5	2.2
C_2H_6	2.3	2.5	2.4	2.1
C_3H_8	1.8	1.8	1.8	1.6
C_4H_{10}	2.6	3.3	3.2	3.1
CO	0.0	0.0	0.0	0.0
H_2S	2.3	1.4	1.3	2.0
H_2O	10.3	6.8	7.1	6.4
NH_3	1.0	0.9	0.8	0.7
Oil	54.6	45.5	47.4	38.9
Asphaltenes	13.3	19.7	19.0	23.3
Preasphaltenes	5.0	8.8	8.1	12.7
Unreacted Coal	4.7	6.7	6.4	7.0
D. H_2 CONSUMPTION, WT. % maf COAL	7.7	6.8	6.7	6.5

OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9008	9009
COAL TESTED	DONKIN HR-9	DONKIN HR-9
A. OPERATING CONDITIONS		
Unit back pressure, MPa	17.24	17.24
Temperatures, °C:		
First-Stage Reactor	400	400
Second-Stage Reactor	425	440
Hot Separator	300	300
Feedrates:		
Slurry, kg/hr	1.012	1.031
Raw Coal, kg/hr	0.283	0.289
Hydrogen, L/hr	636.	636.
SHSV, hr^{-1}; First-Stage	1.00	1.00
Second-Stage	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	20.7	20.3
Catalyst Age, kg coal/kg catalyst		
First-Stage: Harshaw/Filtrol 2	30	61
Second-Stage: Amocat 1C	105	114
B. CONVERSIONS OF maf COAL, WT. %		
Pentane Solubles plus Gases	60.2	65.5
Toluene Solubles plus Gases	80.9	85.9
THF Solubles plus Gases	93.2	93.4
C. PRODUCT YIELDS ON maf COAL, WT. %		
CH_4	2.1	3.9
C_2H_6	1.9	2.4
C_3H_8	1.5	2.0
C_4H_{10}	2.8	1.4
CO	0.0	0.0
H_2S	2.2	1.7
H_2O	6.6	7.0
NH_3	0.8	0.8
Oil	42.3	46.3
Asphaltenes	20.7	20.4
Preasphaltenes	12.3	7.5
Unreacted Coal	6.8	6.6
D. H_2 CONSUMPTION, WT. % maf COAL	7.1	7.2

OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO. COAL TESTED	9010 LINGAN HR-9	9011 DONKIN HR-9	9012 DONKIN HR-9	9014 DONKIN HR-9
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	17.24	17.24	17.24
Temperatures, °C:				
First-Stage Reactor	415	400	420	440
Second-Stage Reactor	425	425	425	425
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.536	1.025	1.037	1.020
Raw Coal, kg/hr	0.430	0.287	0.290	0.286
Hydrogen, L/hr	690.	636.	642.	642.
SHSV, hr^{-1}; First-Stage	0.75	0.50	0.50	0.50
Second-Stage	0.50	0.33	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	14.6	20.5	20.3	20.7
Catalyst Age, kg coal/kg catalyst				
First-Stage: Amocat 1C	47	60	73	86
Second-Stage: Amocat 1C	31	40	49	58
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	62.8	78.2	81.4	85.7
Toluene Solubles plus Gases	82.3	89.8	91.2	93.2
THF Solubles plus Gases	91.4	93.9	93.9	94.8
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.0	2.2	3.1	3.6
C_2H_6	1.9	2.5	3.2	4.2
C_3H_8	1.3	1.8	2.3	3.1
C_4H_{10}	1.8	2.5	2.6	3.7
CO	0.0	0.0	0.0	0.0
H_2S	1.2	2.0	1.8	1.8
H_2O	5.1	8.0	8.0	8.9
NH_3	0.9	1.2	1.3	1.4
Oil	48.6	58.0	59.1	59.0
Asphaltenes	19.5	11.6	9.8	7.5
Preasphaltenes	9.1	4.1	2.7	1.6
Unreacted Coal	8.6	6.1	6.1	5.2
D. H_2 CONSUMPTION, WT. % maf COAL	7.1	8.3	9.7	10.5

OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9015	9016	9017	9018
COAL TESTED	DONKIN HR-9	ILLINOIS NO. 6	LINGAN HR-9	PRINCE HB-9
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	17.24	17.24	17.24
Temperatures, °C:				
First-Stage Reactor	420	400	440	420
Second-Stage Reactor	440	425	425	425
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.008	1.038	1.019	1.043
Raw Coal, kg/hr	0.282	0.291	0.285	0.292
Hydrogen, L/hr	642.	636.	648.	630.
SHSV, hr^{-1}; First-Stage	0.50	0.50	0.50	0.50
Second-Stage	0.33	0.33	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	20.9	20.4	20.7	20.1
Catalyst Age, kg coal/kg catalyst				
First-Stage: Amocat 1C	99	112	125	138
Second-Stage: Amocat 1C	67	7[illegible]	85	94
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	83.0	84.3	77.0	77.7
Toluene Solubles plus Gases	91.6	92.2	89.0	88.9
THF Solubles plus Gases	94.4	95.3	93.6	93.1
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	3.6	2.0	3.8	2.7
C_2H_6	4.2	2.3	4.2	3.1
C_3H_8	3.3	1.8	3.1	2.4
C_4H_{10}	3.5	2.5	3.8	3.0
CO	0.0	0.2	0.0	0.2
H_2S	1.9	2.6	1.3	2.1
H_2O	8.1	11.2	5.8	8.5
NH_3	1.3	1.2	1.2	1.2
Oil	57.1	60.5	53.8	54.5
Asphaltenes	8.6	7.9	12.0	11.2
Preasphaltenes	2.8	3.1	4.6	4.2
Unreacted Coal	5.6	4.7	6.4	6.9
D. H_2 CONSUMPTION, WT. % maf COAL	9.8	9.9	9.7	8.1

OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO. COAL TESTED	9019 DONKIN HR-9	9020 LINGAN HR-9	9022A ILLINOIS NO.6	9022B ILLINOIS NO. 6
A. OPERATING CONDITIONS				
Unit back pressure, MPa	17.24	17.24	17.24	17.24
Temperatures, °C:				
First-Stage Reactor	420	420	400	420
Second-Stage Reactor	425	425	425	425
Hot Separator	300	300	300	300
Feedrates:				
Slurry, kg/hr	1.024	1.033	1.054	1.034
Raw Coal, kg/hr	0.287	0.289	0.295	0.290
Hydrogen, L/hr	642.	648.	636.	636.
SHSV, hr^{-1}; First-Stage	0.50	0.50	0.50	0.50
Second-Stage	0.33	0.33	0.33	0.33
Hydrogen Treatrate, Wt. % Dry Coal	20.6	20.4	20.1	20.4
Catalyst Age, kg coal/kg catalyst				
First-Stage: Amocat 1C	151	164	191	197
Second-Stage: Amocat 1C	103	112	131	135
B. CONVERSIONS OF maf COAL, WT. %				
Pentane Solubles plus Gases	73.0	65.8	77.5	84.9
Toluene Solubles plus Gases	87.5	83.5	89.7	92.9
THF Solubles plus Gases	94.0	92.6	95.2	95.9
C. PRODUCT YIELDS ON maf COAL, WT. %				
CH_4	2.8	2.8	2.0	2.5
C_2H_6	3.0	2.9	2.4	2.9
C_3H_8	2.2	2.2	1.8	2.3
C_4H_{10}	3.1	3.3	2.7	3.3
CO	0.0	0.1	0.2	0.3
H_2S	1.8	1.3	2.4	2.6
H_2O	7.8	4.8	9.7	10.4
NH_3	1.1	1.0	1.1	1.3
Oil	51.2	47.4	55.2	59.3
Asphaltenes	14.5	17.7	12.2	8.0
Preasphaltenes	6.5	9.1	5.5	3.0
Unreacted Coal	6.0	7.4	4.8	4.1
D. H_2 CONSUMPTION, WT. % maf COAL	8.5	8.0	8.9	10.1

APPENDIX C

MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS

MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9000	9001	9002	9003
CATALYST TESTED: First-Stage	HARSHAW/FILTROL 2	HARSHAW/FILTROL 2	HARSHAW/FILTROL 2	HARSHAW/FILTROL 2
Second-Stage	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C
COAL TESTED	LINGAN HR-7	PRINCE HB-9	DONKIN HR-9	LINGAN HR-9
MOISTURE, WT. %	1.28	3.19	2.51	1.08
ASH, WT. % DRY BASIS	3.42	7.16	5.65	4.18
HOURS ON STREAM	24.0	8.8	10.1	10.1
DURATION OF BALANCE PERIOD, HRS.	9.0	2.0	2.0	2.0
INPUT (g):				
H_2O	50.6	19.2	14.9	6.4
Ash	133.4	41.7	32.6	24.5
maf Coal	3768.	542.	545.	563.
Solvent	10161.	1550.	1523.	1526.
H_2	466.6	112.4	115.6	118.4
Total	14580.	2265.	2231.	2238.
OUTPUT (g):				
H_2O	266.6	61.8	54.8	29.3
Ash	115.7	33.0	25.0	16.4
Organic THF Insolubles[1]	281.4	43.5	40.9	54.5
THF Solubles[2]	13177.	1990.	1949.	1951.
H_2	251.8	74.8	79.4	84.2
Other Gases	300.7	65.4	57.3	56.5
Total	14393.	2269.	2206.	2192.
TOTAL MATERIAL RECOVERY, WT. %	98.7	100.1	98.9	97.9

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9004	9005	9006	9007
CATALYST TESTED: First-Stage	HARSHAW/FILTROL 2	HARSHAW/FILTROL 2	HARSHAW/FILTROL 2	HARSHAW/FILTROL 2
Second-Stage	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C
COAL TESTED	ILLINOIS NO. 6	DONKIN HR-9	DONKIN HR-9	DONKIN HR-9
MOISTURE, WT. %	2.91	2.51	2.51	2.51
ASH, WT. % DRY BASIS	9.81	5.65	5.65	5.65
HOURS ON STREAM	10.0	10.1	10.2	10.0
DURATION OF BALANCE PERIOD, HRS.	2.0	2.0	2.0	2.0
INPUT (g):				
H_2O	16.7	14.9	14.9	14.4
Ash	54.7	32.6	32.6	31.6
maf Coal	504.	545.	545.	528.
Solvent	1480.	1523.	1523.	1476.
H_2	114.5	114.5	115.6	113.3
Total	2170.	2230.	2231.	2163.
OUTPUT (g):				
H_2O	68.7	52.0	53.8	48.0
Ash	45.0	22.9	22.1	21.7
Organic THF Insolubles[1]	23.8	36.5	34.7	37.0
THF Solubles[2]	1843.	1930.	1921.	1878.
H_2	75.5	77.4	79.2	79.1
Other Gases	60.6	68.2	66.5	62.0
Total	2117.	2187.	2177.	2126.
TOTAL MATERIAL RECOVERY, WT. %	97.5	98.1	97.6	98.3

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9008	9009	9010	9011
CATALYST TESTED: First-Stage	HARSHAW/FILTROL 2	HARSHAW/FILTROL 2	AMOCAT 1C	AMOCAT 1C
Second-Stage	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C
COAL TESTED	DONKIN HR-9	DONKIN HR-9	LINGAN HR-9	DONKIN HR-9
MOISTURE, WT. %	2.51	2.51	1.08	2.51
ASH, WT. % DRY BASIS	5.65	5.65	4.18	5.65
HOURS ON STREAM	10.02	10.28	24.00	10.03
DURATION OF BALANCE PERIOD, HRS.	2.02	2.23	5.58	1.98
INPUT (g):				
H_2O	14.4	16.2	25.7	14.3
Ash	31.5	35.5	99.2	31.3
maf Coal	527.	592.	2275.	523.
Solvent	1472.	1656.	6170.	1461.
H_2	115.6	127.6	346.5	113.3
Total	2161.	2427.	8916.	2143.
OUTPUT (g):				
H_2O	49.1	57.5	141.5	56.1
Ash	21.0	25.4	84.8	21.1
Organic THF Insolubles[1]	36.0	39.0	196.6	31.7
THF Solubles[2]	1870.	2064.	8019.	1823.
H_2	78.3	85.0	184.0	69.7
Other Gases	60.0	72.3	207.4	63.4
Total	2114.	2343.	8833.	2065.
TOTAL MATERIAL RECOVERY, WT. %	97.9	96.5	99.1	96.4

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9012	9014	9015	9016
CATALYST TESTED: First-Stage	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C
Second-Stage	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C
COAL TESTED	DONKIN HR-9	DONKIN HR-9	DONKIN HR-9	ILLINOIS NO. 6
MOISTURE, WT. %	2.10	2.10	2.10	3.20
ASH, WT. % DRY BASIS	5.63	5.63	5.63	9.81
HOURS ON STREAM	10.08	10.18	10.03	10.07
DURATION OF BALANCE PERIOD, HRS.	2.03	2.02	2.00	2.00
INPUT (g):				
H_2O	12.4	12.1	11.8	18.6
Ash	32.5	31.8	31.1	55.2
maf Coal	545.	533.	521.	507.
Solvent	1516.	1483.	1451.	1494.
H_2	117.3	116.7	115.6	114.5
Total	2223.	2177.	2131.	2189.
OUTPUT (g):				
H_2O	56.1	59.6	54.2	75.4
Ash	22.7	18.3	19.4	47.9
Organic THF Insolubles[1]	33.3	28.0	29.3	23.8
THF Solubles[2]	1897.	1790.	1798.	1878.
H_2	64.5	60.7	64.6	64.5
Other Gases	78.6	95.2	92.7	64.1
Total	2152.	2052.	2058.	2154.
TOTAL MATERIAL RECOVERY, WT. %	96.8	94.3	96.6	98.4

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9017	9018	9019	9020
CATALYST TESTED: First Stage	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C
Second-Stage	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C	AMOCAT 1C
COAL TESTED	LINGAN HR-9	PRINCE HB-9	DONKIN HR-9	LINGAN HR-9
MOISTURE, WT. %	1.08	3.19	2.10	1.08
ASH, WT. % DRY BASIS	4.18	7.16	5.63	4.18
HOURS ON STREAM	10.08	10.03	10.11	10.07
DURATION OF BALANCE PERIOD, HRS.	2.05	2.00	2.03	1.98
INPUT (g):				
H_2O	6.3	18.6	12.2	6.2
Ash	24.2	40.5	32.1	23.7
maf Coal	554.	525.	538.	543.
Solvent	1504.	1501.	1497.	1472.
H_2	119.6	113.4	117.3	115.5
Total	2208.	2199.	2197.	2160.
OUTPUT (g):				
H_2O	38.4	63.4	53.9	32.4
Ash	13.9	30.6	21.7	15.1
Organic THF Insolubles[1]	35.6	36.2	32.3	40.2
THF Solubles[2]	1891.	1850.	1914.	1874.
H_2	65.7	71.1	71.5	71.8
Other Gases	96.8	77.1	75.7	73.4
Total	2141.	2128.	2169.	2107.
TOTAL MATERIAL RECOVERY, WT. %	97.0	96.8	98.7	97.5

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

RUN NO.	9022A	9022B
CATALYST TESTED: First-Stage	AMOCAT 1C	AMOCAT 1C
Second-Stage	AMOCAT 1C	AMOCAT 1C
COAL TESTED	ILLINOIS NO. 6	ILLINOIS NO. 6
MOISTURE, WT. %	3.31	3.31
ASH, WT. % DRY BASIS	9.81	9.81
HOURS ON STREAM	10.10	14.76
DURATION OF BALANCE PERIOD. HRS.	2.03	2.03
INPUT (g):		
H_2O	19.8	19.5
Ash	56.8	55.8
maf Coal	523.	513.
Solvent	1541.	1512.
H_2	116.2	116.2
Total	2257.	2217.
OUTPUT (g):		
H_2O	70.3	72.8
Ash	47.0	45.0
Organic THF Insolubles[1]	25.0	20.9
THF Solubles[2]	1924.	1875.
H_2	69.7	64.2
Other Gases	66.2	77.4
Total	2202.	2155.
TOTAL MATERIAL RECOVERY, WT. %	97.6	97.2

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

APPENDIX D

RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

APPENDIX D

RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

In this study, high performance liquid chromatography (HPLC) was used to separate coal liquids into saturates, aromatics and polar compounds. Details pertaining to the HPLC method have been given in an earlier report (6).

Aromatics are separated into "double bond" fractions using this method. A number of model compounds were used to define the specific regions of the chromatograph as shown in Figure 7. The regions were defined by the number of double bonds in a given hydrocarbon molecule ranging from 0 (saturates) to 8 (tetracyclic PAH). Elution profiles for hydrogenated anthracene oil and the pentane soluble fraction of C_5+ liquids from selected two-stage coal liquefaction experiments are shown in Figures 8 - 17. The C_5+ liquids were obtained by combining separator overheads light oil and separator bottoms liquid recovered by vacuum filtration at 75°C.

Results of HPLC analysis presented in Table 14 show that pentane soluble oil from Run Nos. 9001 - 9004 contain more saturates and less aromatics than the reaction solvent HAO 91, as do Run Nos. 9011, 9016, 9018 and 9019 compared to HAO 92. Furthermore, the pentane soluble fractions contain a higher level of polar compounds than HAO 91 and HAO 92.

It was also noted that concentrations of 5, 6 and 7 'double bond' aromatics were lower in the pentane soluble fractions from Run Nos. 9002, 9003 and 9004 than in the reaction solvent HAO 91. This trend continued in the pentane soluble fractions from Run Nos. 9011, 9016, 9018 and 9019.

Figure 7 - HPLC ELUTION PROFILES FOR MODEL COMPOUNDS

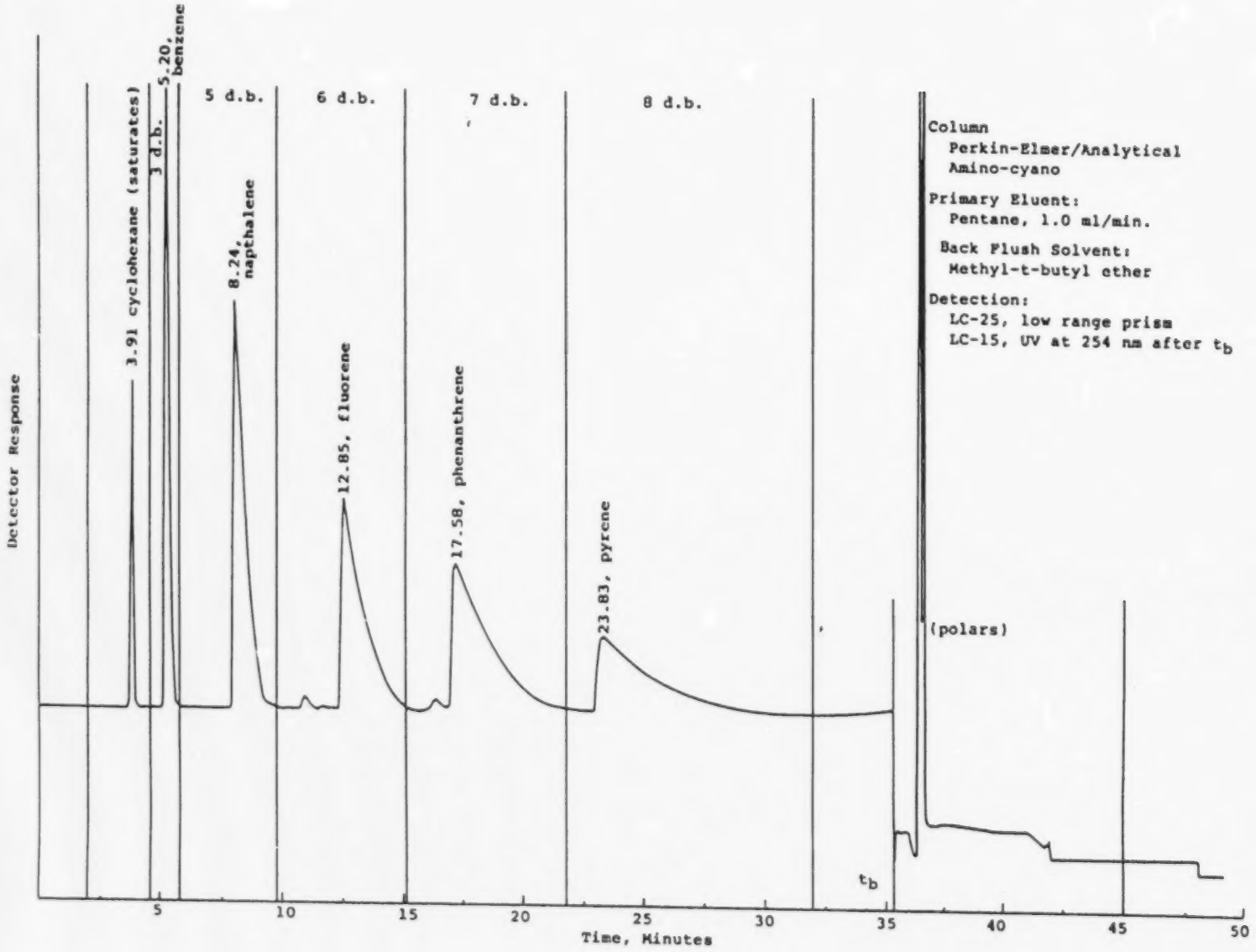


Figure 8 - HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL, BATCH HAO 91

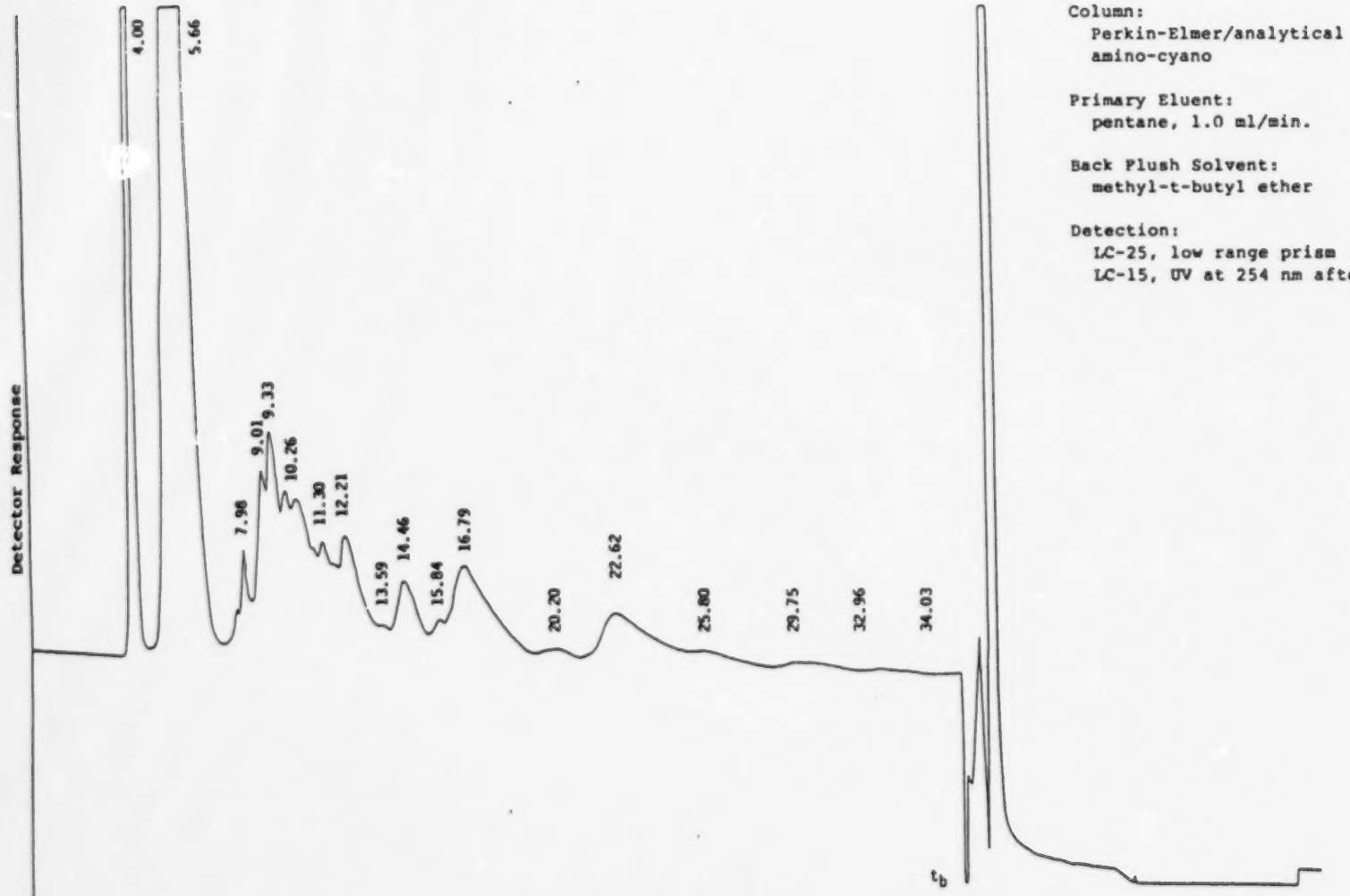


Figure 9 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9001

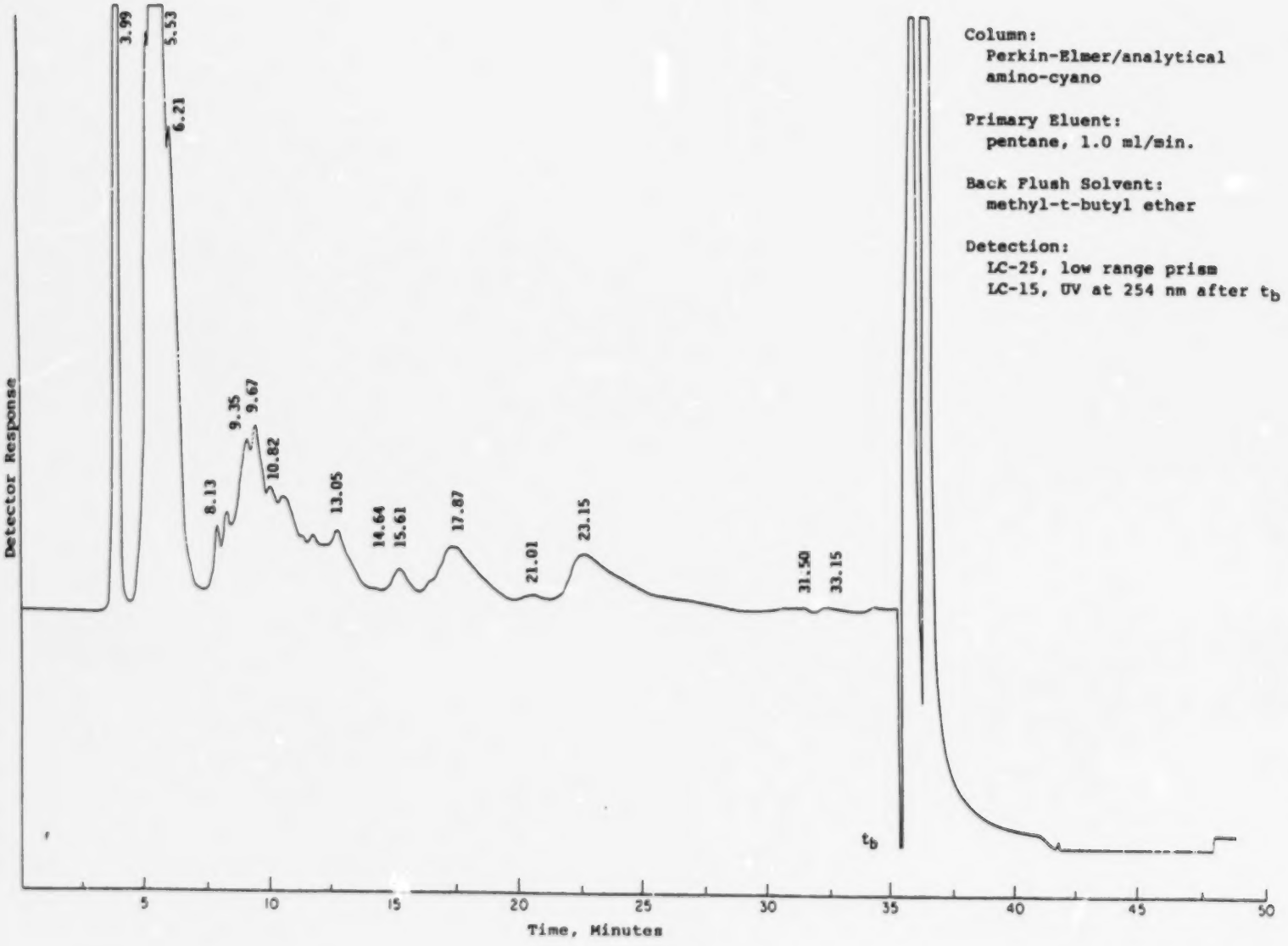


Figure 10 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9002

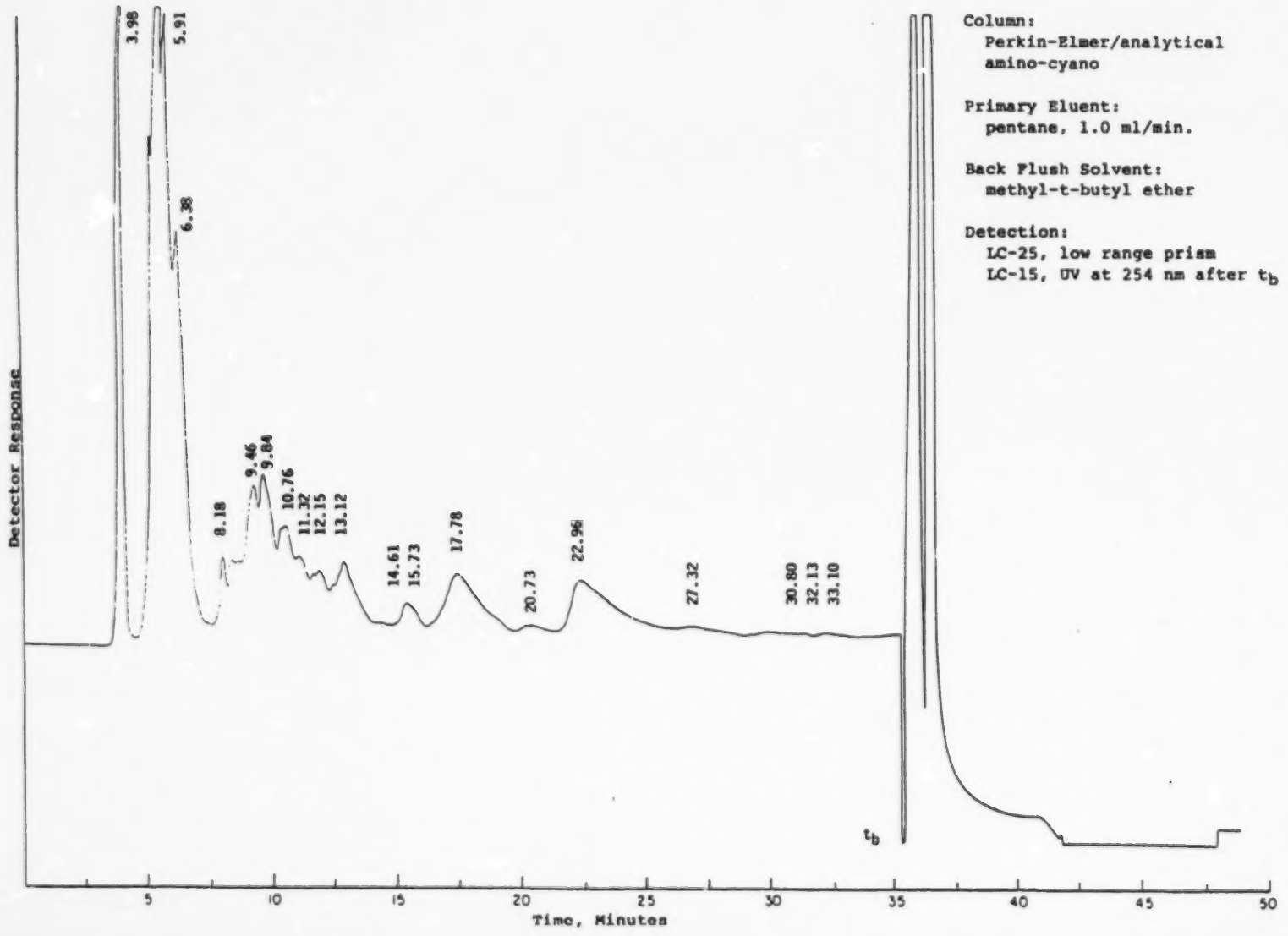
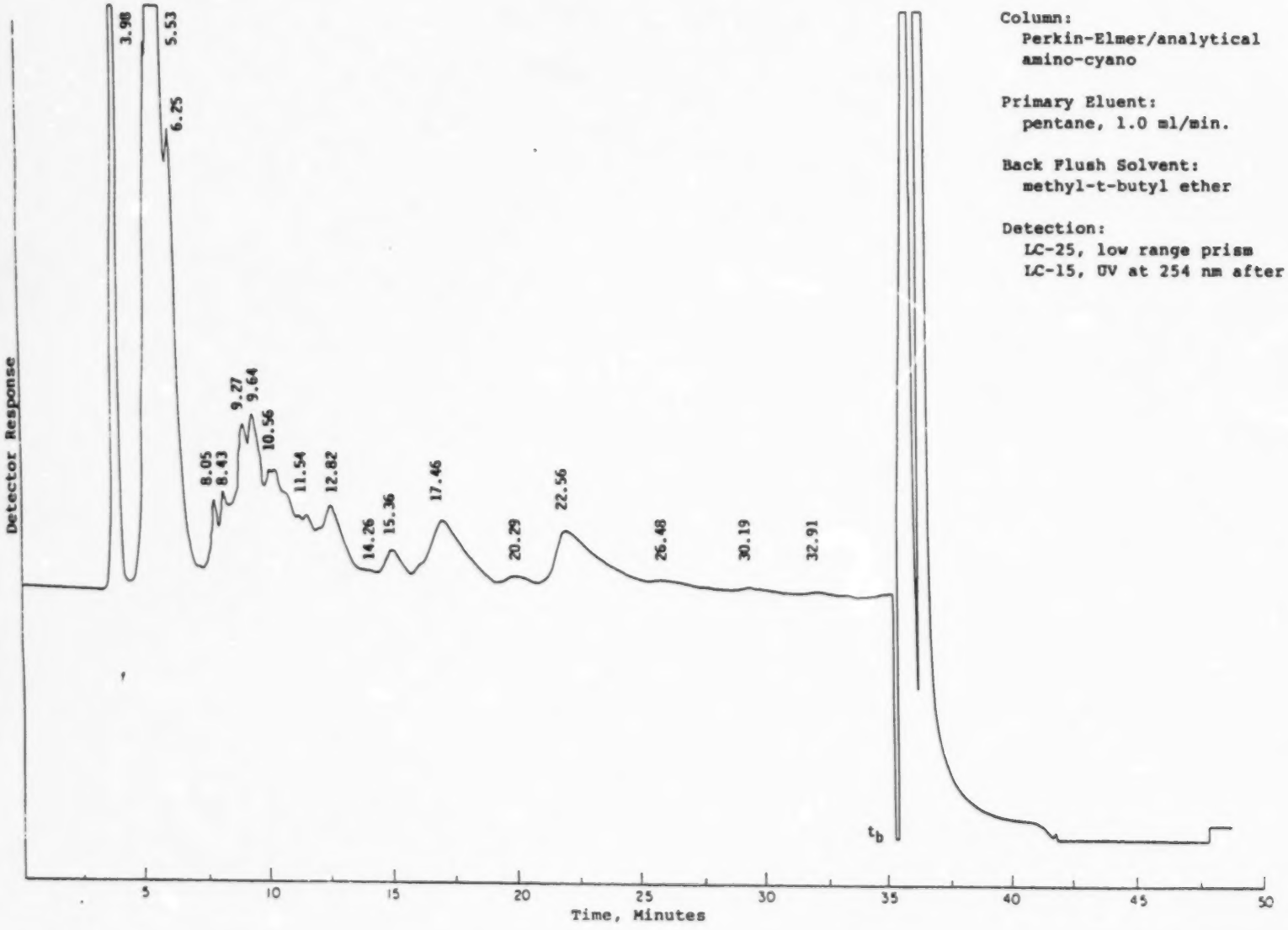


Figure 11 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9003



Column:
Perkin-Elmer/analytical
amino-cyano

Primary Eluent:
pentane, 1.0 ml/min.

Back Flush Solvent:
methyl-t-butyl ether

Detection:
LC-25, low range prism
LC-15, UV at 254 nm after t_b

Figure 12 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9004

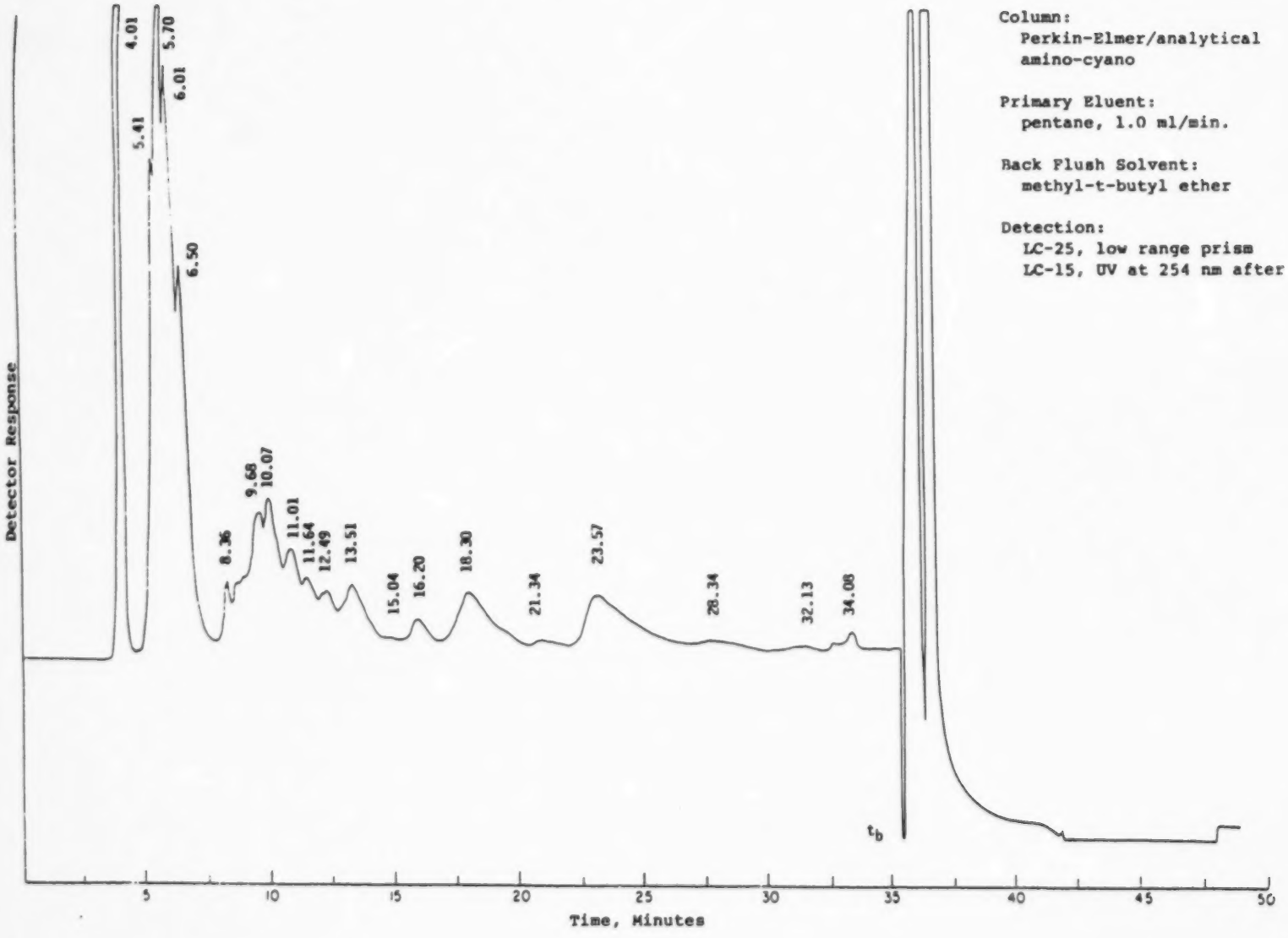


Figure 13 - HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL, BATCH HAO 92

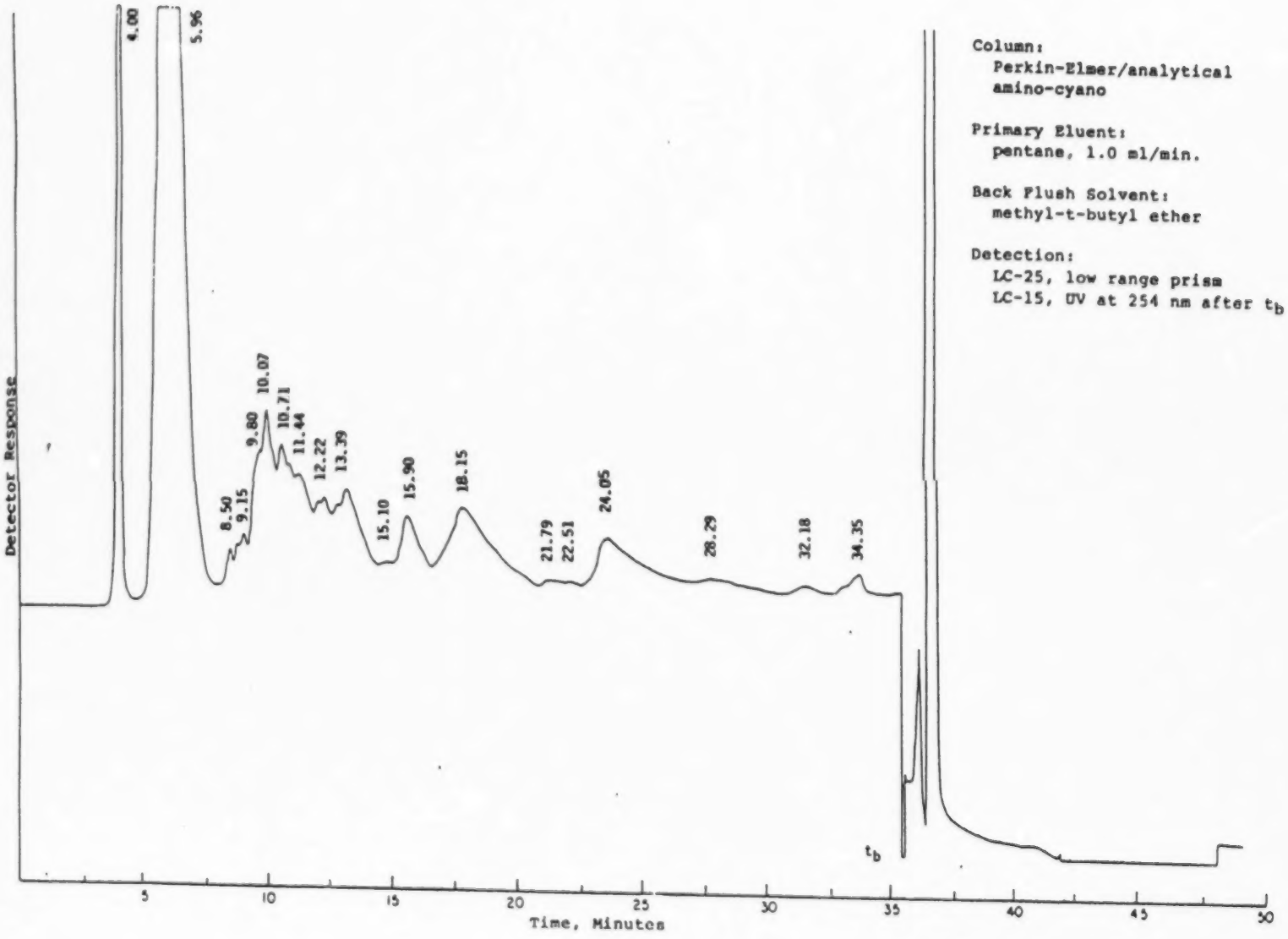


Figure 14 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9011

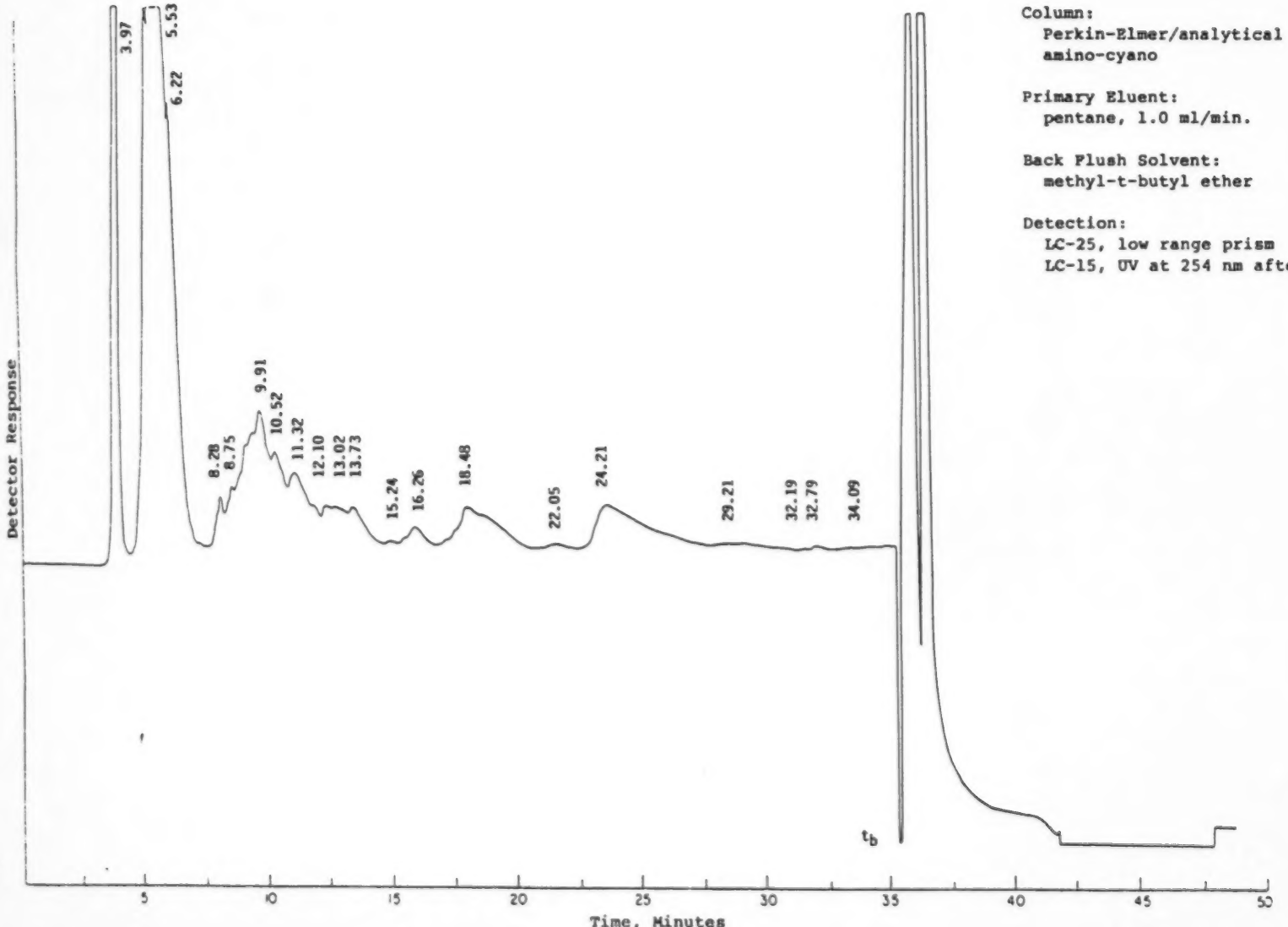


Figure 15 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9016

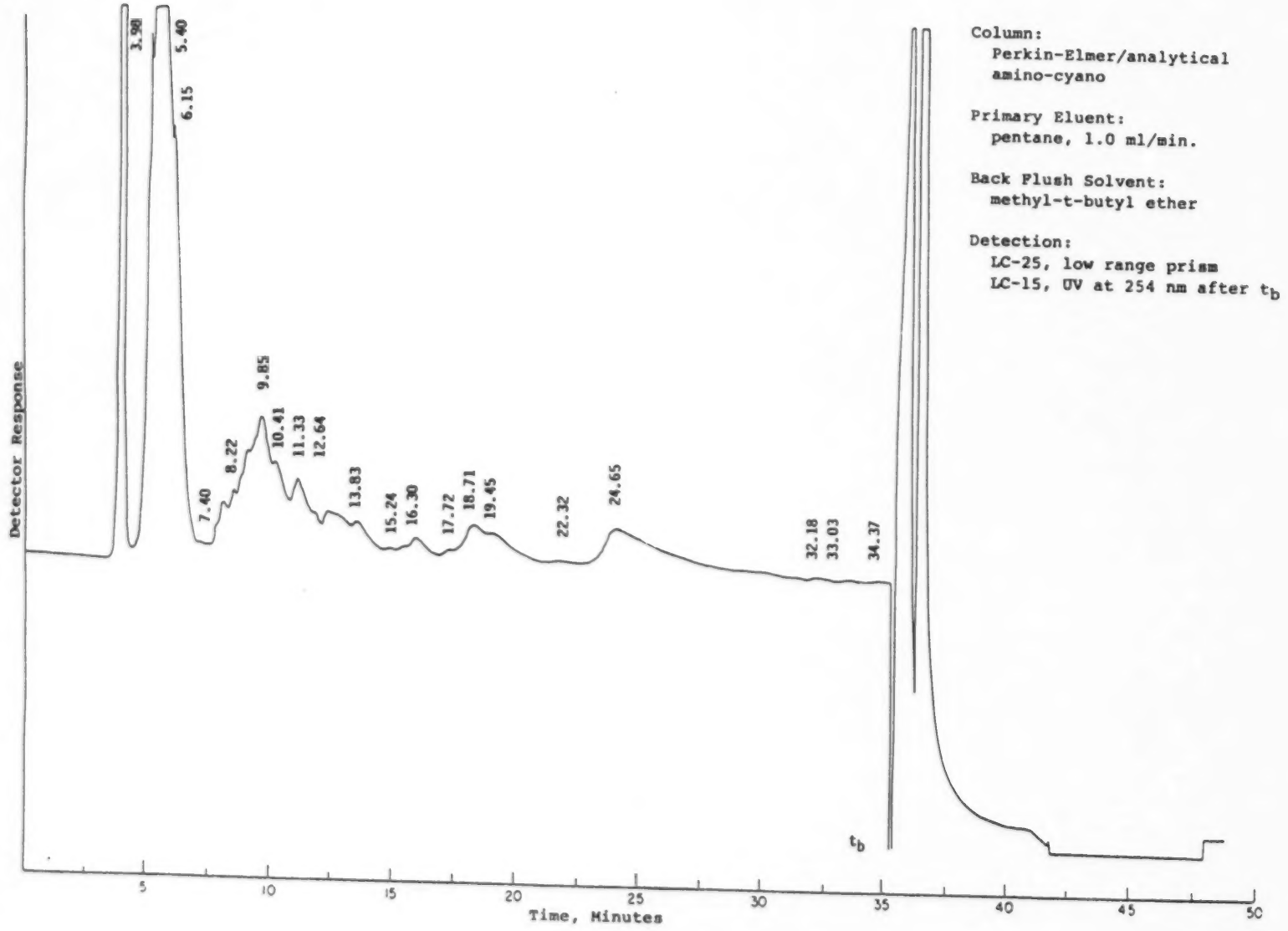


Figure 16 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9018

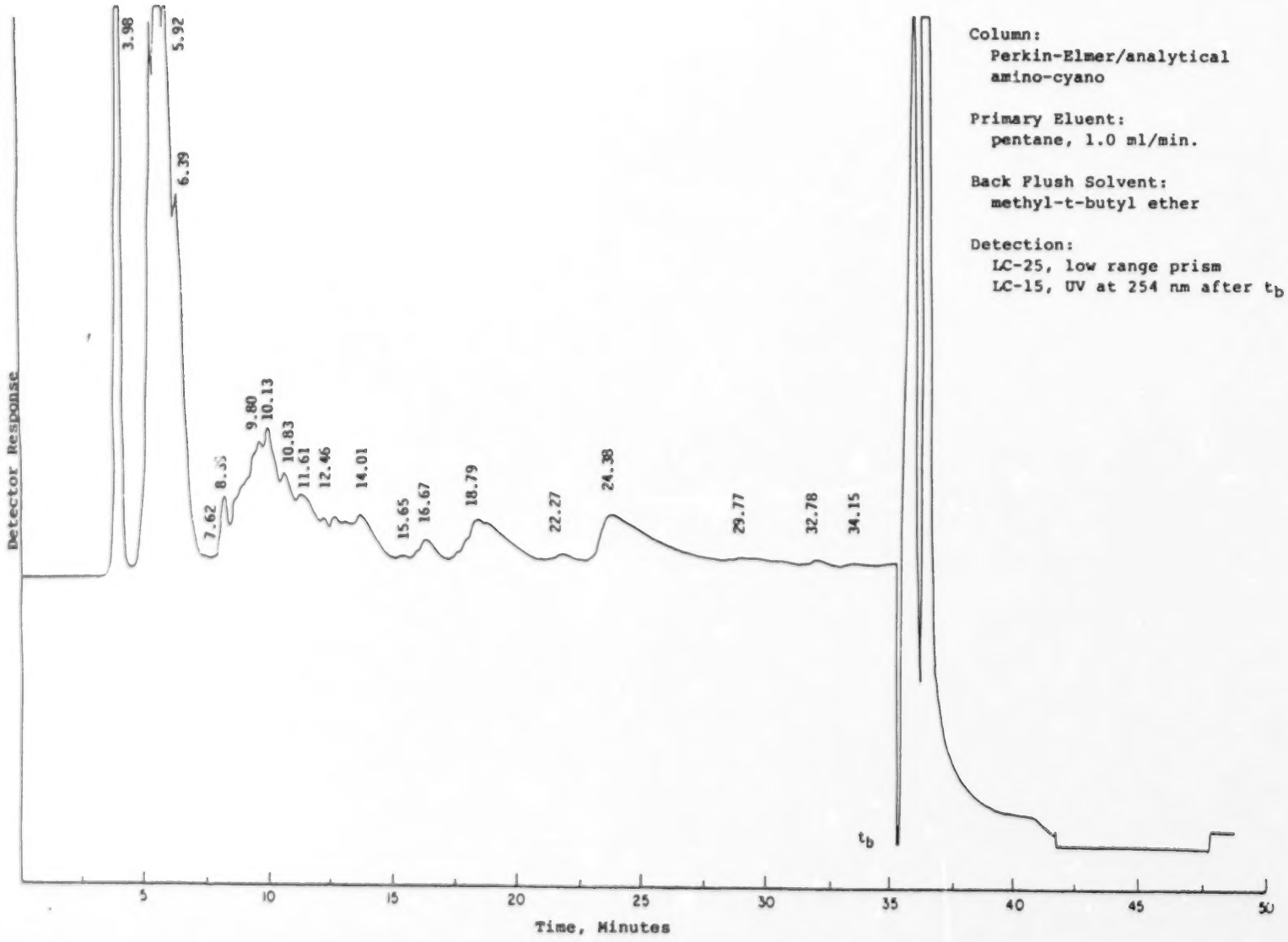


Figure 17 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF C_5^+ LIQUID FROM RUN NO. 9019

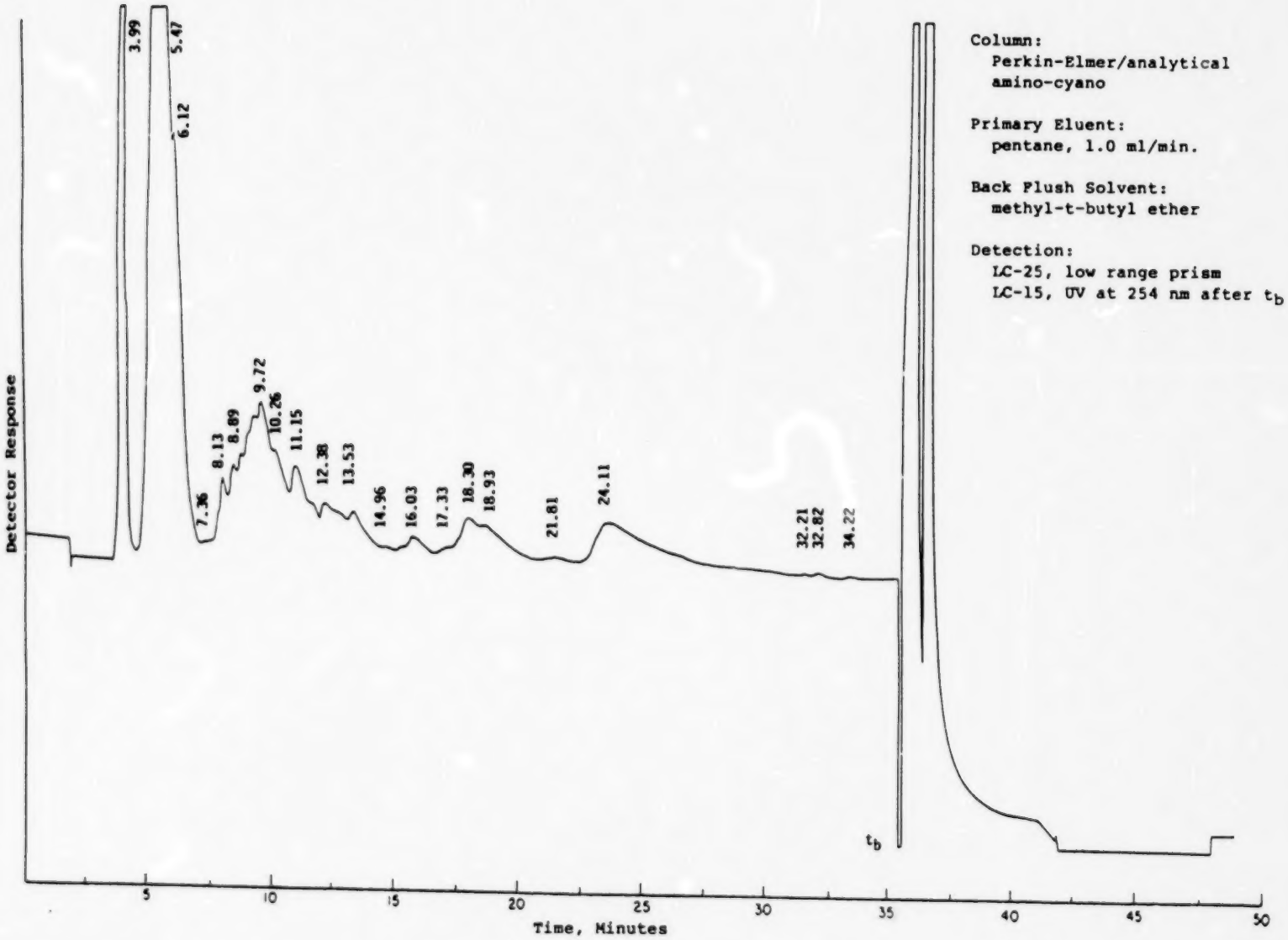


TABLE 14

HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL AND THE PENTANE SOLUBLE FRACTION OF C_5+ LIQUIDS FROM SELECTED COAL LIQUEFACTION EXPERIMENTS

SAMPLE DESCRIPTION:	HAO 91	RUN NO. 9001 PRINCE HB-9	RUN NO. 9002 DONKIN HR-9	RUN NO. 9003 LINGAN HR-9	RUN NO. 9004 ILLINOIS NO. 6
Specific Gravity, 60/60°F	1.0341	1.0218	1.0271	1.0248	1.0091
Carbon, Wt. %	89.89	90.34	90.39	90.49	90.46
Hydrogen, Wt. %	9.09	9.38	9.21	9.32	9.58
Nitrogen, Wt. %	0.16	0.26	0.24	0.29	0.22
Sulphur, Wt. %	0.008	0.051	0.065	0.035	0.055
Pentane Insolubles, Wt. %	nil	4.76	6.00	7.00	3.51
HPLC DATA FOR PENTANE SOLUBLE FRACTION:					
Saturates, Wt. %	14.15	17.91	17.41	17.41	17.73
Aromatics, Wt. %	78.91	65.87	69.61	71.32	69.16
3 Double Bonds	28.78	27.03	26.28	29.01	28.11
5 Double Bonds	19.69	14.86	18.43	16.72	17.72
6 Double Bonds	15.55	10.81	12.28	12.28	9.88
7 Double Bonds	8.27	9.12	5.80	6.14	6.81
8 Double Bonds	6.62	4.05	6.82	7.17	6.64
Polars, Wt. %	6.95	16.22	12.97	11.27	13.12

TABLE 14 (Continued)

HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL AND THE PENTANE SOLUBLE FRACTION OF C_5+ LIQUIDS FROM SELECTED COAL LIQUEFACTION EXPERIMENTS

SAMPLE DESCRIPTION:	HAO 92	RUN NO. 9011 DONKIN HR-9	RUN NO. 9016 ILLINOIS NO. 6	RUN NO. 9018 PRINCE HB-9	RUN NO. 9019 DONKIN HR-9
Specific Gravity, 60/60°F	1.0381	1.0151	1.0101	1.0161	1.0184
Carbon, Wt. %	89.95	88.89	88.36	88.29	88.76
Hydrogen, Wt. %	9.03	9.56	9.76	9.42	9.70
Nitrogen, Wt. %	0.14	0.18	0.17	0.16	0.21
Sulphur, Wt. %	0.009	0.030	0.032	0.031	0.047
Pentane Insolubles, Wt. %	nil	3.47	1.82	3.18	4.10
HPLC DATA FOR PENTANE SOLUBLE FRACTION:					
Saturates, Wt. %	13.86	21.43	22.96	19.26	19.44
Aromatics, Wt. %	81.18	67.33	65.58	70.25	70.43
3 Double Bonds	31.35	28.23	30.13	31.16	29.89
5 Double Bonds	19.14	16.32	13.18	15.01	15.14
6 Double Bonds	14.85	10.54	9.76	11.33	10.30
7 Double Bonds	8.58	5.10	5.65	7.37	7.14
8 Double Bonds	7.26	7.14	6.86	5.38	7.96
Polars, Wt. %	4.95	11.23	11.47	10.48	10.14

APPENDIX E

OPERATING PROCEDURE FOR TWO-STAGE CONTINUOUS COAL LIQUEFACTION UNIT

PREPARATION OF UNIT PRIOR TO ACTUAL OPERATION

This procedure is normally carried out the day before a run.

1. Open valves 1, 7, 11, 14, 15, 116, 119 and 122.
2. Depressure the system to 345 kPa by opening valves 17 and 18. Close valves 17 and 18.
3. Open valve 2 and valve 8 and pressurize the system as well as the 7.6 liter emergency blowdown vessel to 4.14 MPa. Close valve 8 and continue pressurizing the system until the pressure is at the desired level either 10.34 MPa or 13.79 MPa. Close valve 2.
4. Leave for a few hours and monitor pressure.
5. Set the forward pressure regulator and the back pressure regulator to the desired settings.
6. Ensure that alarm switch No. 14 is deactivated.

ACTUAL OPERATION

Start-Up

1. Turn on the cooling water to the magnedrive stirrers.
2. Set the controllers at the desired settings for both the first-stage and second-stage reactors.

	Stage 1	Stage 2
reactor controller, °C	343	288
furnace controller, °C	343	316
zone controllers, %	60	100

3. Turn on the temperature controller for high pressure separator No. 1, V-5, at 300°C.
4. Turn on the temperature controller for high pressure separator No. 2, V-6, at 300°C.
5. Gradually increase the furnace controllers for stage 1 and stage 2 to the desired levels. NEVER allow the heating rate of the reactors to exceed 120°C per hour.

6. After two hours of heat-up time follow steps 7, 8 and 9.

7. Turn on heating tapes on slurry transfer lines:

   No. 1 - 33%, at start of run gradually increase to 100%

   No. 2 - 33%, at start of run gradually increase to 66%

   No. 3 - 33%

   No. 4 - 33%

8. Turn on condenser cooling water.

9. Activate K-Ray electronics for liquid level measurement system.

10. After one hour open the window on the radioactive source.

11. Turn on the hydrogen flow control system fifteen minutes prior to run.

Procedure

a) Turn on the power.
b) Open valve 3.
c) Set the air pressure at 138 kPa.
d) Adjust the setpoint knob until the ball is in the center, then switch from manual to automatic.
e) Set the pen to the desired level.
f) Open valve 123 and close valve 122.
g) Allow the hydrogen flow control system to stabilize.
h) Monitor exit gas flow rate for 15 minutes.

12. Purge the feed tank with nitrogen 15 minutes prior to slurry addition. Maintain a small nitrogen flow throughout the run.

13. Charge V-1 with the slurry. Open valve 101 and turn on the circulating pump.

14. Turn off the circulating pump for 15 seconds and then turn the circulating pump back on. This fills void spaces or gas pockets in the pump with slurry and ensures proper weight readings on the toledo scale. Turn on the alarm switch No. 14 in the side panel.

15. When the temperatures of the reactors are at the desired levels open valve 102 and purge the high pressure metering pump with slurry until the liquid is gas free.

Run

16. Open the plant valve (valve 5 or 6) and activate the high pressure metering pump.

17. Turn on the V-3, V-4 and V-5 magnedrive stirrers.

18. Turn on the cooling fan for the magnedrive stirrer on V-4.

19. Monitor the pressures, temperatures, slurry feedrate and vent gas flow rates through the run.

20. A rapid increase in the reactor 1 temperature will occur 10 to 20 minutes after the high pressure metering pump has been activated. When the reactor temperature reaches 4°C below the target temperature turn the furnace temperature controller down until the red light goes off. Increase the furnace temperature controller back to the original setpoint when reactor 1 temperature levels off.

21. Open valves 111, 107, 104 and 105 and purge V-9 with nitrogen, 10 minutes prior to step 22.

22. After 2.3 kg of slurry has been pumped, open valve 22.

23. Set timer K-5 for the automatic letdown (ALD) system to maintain a constant level in V-5.

24. Open valve 19.

25. Set the selector switch on "Automatic During High Alert" and activate the automatic letdown system.

26. After the first ALD cycle (mark this on the data sheet) close valve 105.

27. Sample vent gas streams 15 minutes prior to the steady state sampling period.

28. Drain liquid from the separator overheads knock-out (V-7) periodically and 5 minutes prior to the steady state sampling period by opening and closing valves 17 and 18 sequentially.

29. To initiate the steady state sampling period, record the toledo scale weight after valve "A" has closed on the final ALD cycle of the slop period.

30. Record the wet test meter reading and the circulating and metering pump pan weights.

31. Open valves 108 and 23. Close valves 22 and 107. Open valve 106 and start a nitrogen purge.

32. On the next ALD cycle, the first in the steady state sampling period, mark the stroke made on the hydrogen flow recorder.

33. Close valve 106.

34. Open valve 120 and drain V-15. Close valve 120.

35. Sample vent gas streams 15 minutes prior to the end of the steady state sampling period.

36. Five minutes prior to end of run, drain liquid from separator overheads knock-out, V-7, by opening and closing valves 17 and 18 sequentially.

37. At the end of the period, record the toledo scale weight after valve "A" has closed on the final ALD cycle of the sampling period.

38. Open valves 107 and 22. Close valves 23 and 108. Open valve 105 and start a nitrogen purge. After the next ALD cycle close valve 105.

39. Record the wet test meter reading and the circulating and metering pump pan weights. Empty contents of V-15.

40. Turn the reactor controller setpoints on stage 1 and stage 2 reactors down to 260°C. Turn the furnace controller setpoints on stage 1 and stage 2 reactors down to 204°C. Reduce voltage to heating tapes to 33% on slurry transfer lines. Turn off high pressure separator heaters.

41. Record the toledo scale weight and deactivate the circulating pump for 15 seconds. Reactivate the circulating pump and record the toledo scale weight.

42. Turn down the hydrogen flow control system to ca. 100 S/D.

43. Deactivate the high pressure metering pump.

44. Close the plant valve (valve 5 or 6) and valves 11 and 14.

45. Deactivate the circulating pump and drain slurry from the feed tank.

46. Close valves 101 and 102 and add flush oil to V-1, open valve 101 and turn on the circulating pump.

Flush Cycle and Shutdown

47. Open the priming valve above the metering pump and allow 1 kg of flush oil to exit. Close the priming valve.

48. Open the plant valve and activate the high pressure metering pump.

49. Reactivate the reactor controller setpoints in both reactors to 371°C. Reactivate the furnace controller setpoints in both reactors to ca. 540°C. Turn on the heater on separator No. 1 to 300°C. Increase voltage on line heaters to 66% for the preheater and 33% for the interstage and slurry transfer lines. Increase the hydrogen flow control setpoint to 300 S/D.

50. Note: Keep temperatures of stage 1 and stage 2 reactors between 343°C and 371°C during flush cycle.

51. Open valves 111 and 120 and drain V-15. Close valves 120 and 111.

52. When approximately 3.0 kg of oil has been pumped into V-5, open valves 111, 108, 23 and 19. Activate the ALD system allowing it to operate until at least 14 kg of flush oil has been pumped. Close valve 19, allow one more cycle. Deactivate the ALD system and close valves 23, 108 and 111.

53. Pull the plug out for electronics on the ALD system and close the window on the radioactive source. Shut off hydrogen control system.

Procedure

a) Turn the regulator knob until the ball is in the centre.
b) Switch from automatic to manual.
c) Shut off the air flow.
d) Close valve 3.
e) Turn off the power

54. Deactivate the metering pump.

55. Shut off all heating tapes, heaters and the reactor furnaces.

56. Turn off the condenser cooling water. Leave the magnedrive water on overnight.

57. Close the plant valve and valves 11 and 14.

58. Withdraw oil from V-5.

59. Open valves 14 and 15 to equalize the pressure in V-5, V-6 and V-7.

60. Open valve 11 to equalize the pressure throughout the system.

61. Open valve 18 and slowly open valve 17 to release system pressure to 11.03 MPa. Close valve 17 and then valve 18.

62. Close the interstage valve and valve 11.

63. Transfer contents of V-4 into the 7.6 liter high pressure vessel by opening valve 9. Let stand for 5 minutes then close valve 9. Ensure that pressure in V-4 stays above 6.89 MPa.

64. Transfer contents of V-3 into the 7.6 liter high pressure vessel by opening valve 8. Let drain for 5 minutes.

65. Open valve 11 and equalize pressure in V-4, V-5 and V-6.

66. Open valve 2 and add high pressure hydrogen to V-3 until its pressure is 689 kPa in excess of V-4. Close valve 2.

67. Open the interstage valve (valve 7) to equalize system pressure.

68. Open valve 2 and add hydrogen to the system until the pressure is 11.38 MPa.

69. Drain flush oil from V-1.

70. Turn off electronics for the toledo scale and digital pressure meter.

71. Shut off alarm switch No. 14 in the side panel.

POST RUN OPERATIONS

This procedure is normally carried out the day after the run.

1. Open valve 2 and pressurize the system to 10.34 MPa.

2. Close the interstage valve (valve No. 7) and valves 11 and 14.

3. Depressure the 7.6 liter pressure vessel to 4.83 MPa.

4. Drain contents of V-5 by opening valves 110, 26 and 24. Close valves 24, 26 and 110.

5. Open valve 14 and equalize the pressure in V-5, V-6 and V-7.

6. Empty contents of V-4 by opening valve 9. Close valve 9.

7. Depressure the 7.6 liter pressure vessel to 4.83 MPa.

8. Empty contents of V-3 by opening valve 8. Close valve 8.

9. Open valve 11.

10. Follow steps 66 and 67. Then open valve 2 and add hydrogen to the system until the pressure is ca. 12.93 MPa.

11. Depressure the 7.6 liter pressure vessel to 1.38 MPa. Empty contents of the 7.6 liter vessel into the 10 liter low pressure vessel.

12. Follow steps 1 to 4 on page E-2.

Figure 18 - SCHEMATIC DIAGRAM OF TWO-STAGE COAL LIQUEFACTION BENCH UNIT